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RARE GAS MEASUREMENTS IN METEORITES
AND POSSIBLE APPLICATIONS TO THE LUNAR SURFACE

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ABSTRACT

In the first part of this paper applications and results obtained from rare gas measurements on meteorites are discussed, showing the usefulness and importance of such measurements. The rare gases in a meteorite originate from five different sources and, thus, a wide range of information on the history of the meteorite and its surroundings can be deduced from rare gas measurements. K-Ar⁴⁰ and U, Th-He⁴ ages determine the time of the meteorite's formation and its subsequent thermal history. Rare gas isotopes such as He³, Ne²¹, and Ar³⁸ produced by cosmic ray induced spallation reactions in the meteorite, date the break up of the meteorite parent bodies into smaller fragments. These spallation produced isotopes may also be used for monitoring the history of the cosmic radiation. Decay products of "short lived" isotopes such as Xe¹²⁹ from the decay of I¹²⁹ measure the time interval between the end of nucleosynthetic processes and the formation and cooling of the meteorite parent bodies. Trapped rare gases, a feature found in certain meteorites, give direct information on the atmosphere of the meteorite parent bodies and probably also of the solar wind in the past.

From these measurements we know that most of the stone meteorites were formed 4.5×10^9 years ago and have not been heated since then. The time interval between the end of certain processes of element synthesis and the formation of the meteorites is probably only of the order of 10^8 years. Most stone meteorites suffered no further disturbance until some 10^7 years ago, when they were broken out of larger meteorite parent bodies.

In the second part of this paper the possible applications of rare gas measurements to lunar surface material are discussed in detail. The time of formation of surface features such as craters, maria etc. can be determined with the K-Ar⁴⁰ or the U, Th-He⁴ method. Thus, together with a relative lunar stratigraphic time scale, the approximate absolute age of many

surface features could be obtained from relatively few absolute age determinations. The time dependence of the crater frequency would then give important clues on the history of small bodies in the solar system. The age of the moon, if such an age is meaningful at all, may of course also be determined.

The lunar surface is exposed directly to the high- and low-energy particle radiation. Particles with high energies will lead to the formation of new isotopes due to nuclear reactions, in most cases spallation reactions. These spallation produced isotopes may be used for determining the age of surface features, erosion and accumulation rates etc. Furthermore, spallation produced isotopes, in particular the rare gases, may serve as monitor of the history of the cosmic radiation. Three different sampling methods for such a study are given and their sensitivity to changes in the cosmic radiation discussed. Lunar material, with its much better known shielding history is of course greatly superior to meteoritic material for such studies. Furthermore, from the mass distribution of spallation products it is possible to deduce the cosmic ray spectrum.

Particles with very low energies, (e.g. solar wind) will be trapped in the immediate surface layer of the moon. Measurements of such trapped gases will yield information on the history of the low energy particle radiation and of its composition. Finally, the problem of diffusion losses of rare gases is discussed.

1. INTRODUCTION

At the present time it may seem rather ambitious to discuss the results which could be obtained from radioactive dating and from rare gas measurements on lunar material. It may still take many years until such samples will be available for analyses. Nevertheless, it seems important to us, to show the vast amount of information that may be gained from rare gas measurements and radioactive dating of lunar material. Thus, the absolute age of surface features, such as craters and maria, might be determined, the absolute age of the moon as a whole body inferred, and the time of separation of the lunar material from general or special nucleosynthetic processes measured. Furthermore, the erosion history of the lunar surface could be deduced from the concentration of spallation rare gases. From these results concerning the moon itself, more general conclusions regarding the history of the solar system might be drawn (UREY 1962).

The measurement of rare gases on lunar material will also be very important for unraveling the history of the particle radiation field in our solar system. Most of our information on the intensity and spectrum of the cosmic radiation in the past has been obtained from measurements on meteorites (HOUTERMANS 1954; ARNOLD 1961; ARNOLD, HONDA and LAL 1961; GEISS, OESCHGER and SCHWARZ 1961; GEISS 1963; ARNOLD and HONDA 1964). The limited results obtained so far are mainly due to the nonavailability of meteorites with a suitable and a known history. However, as will be discussed in this article, lunar surface material keeps a much better record of the history of the cosmic radiation and more information can be obtained from it than from meteorites.

The amount of material required for a rare gas analysis is generally much smaller than one gram. Analyses have been successfully performed on a few milligrams (EBERHARDT, GEISS and GROEGLER 1964). Even

if preliminary mineral separations are required amounts of a few grams should be adequate. Thus, many different samples could be returned from a single lunar mission.

It is not absolutely necessary to return material to earth for rare gas measurements. A system for remote controlled extractions and mass spectrometric analyses on an unmanned lunar mission has been shown to be feasible (EBERHARDT and GEISS 1962, 1963, 1964). Returning the sample to earth has of course the advantage that additional investigations, such as chemical and mineralogical analyses may be performed on it. Also, a mineral separation prior to the rare gas analyses may greatly improve the significance of these measurements.

There exists of course the very slight chance, that some of the meteorites collected on earth may be of lunar origin. (UREY 1959, O'KEEFE 1961). The identification of such material will probably be based on statistical arguments concerning the cosmic ray exposure ages, the primordial rare gas contents, luminescence and similar properties of meteorites and the meteorite classes (EBERHARDT and GEISS 1964 b). But very likely it will not be possible to prove unambiguously a lunar origin without having collected samples for direct comparison. Furthermore, their initial location on the moon will not be known, a serious drawback compared to directly collected material.

Several authors have proposed a lunar origin for tektites. (NININGER 1952; O'KEEFE 1958; VARSAVSKY 1958; GOLD 1958; O'KEEFE 1959, 1961; O'KEEFE and SHUTE 1961; CHAPMAN and LARSON 1963). Many arguments have been brought forward against such a theory (BARNES 1958; KOPAL 1958; UREY 1958; UREY 1962 a, 1963). At the present time, and probably also in the near future, this question will not be decided unequivocally. Even if tektites were of direct lunar origin they would have been heated above their melting point during entry into the earth's atmosphere and thus have lost their rare gases, and probably also some of the other elements, which they formerly might have contained. For this reason they are not suitable

for the studies proposed in this paper.

In the first part of this article a survey of the results obtained from rare gas measurements in meteorites is given. This will show the possible sources of rare gases (radioactive decay, spallation, trapping etc.) in lunar material and the type of general conclusions which can be drawn from their concentrations. In comparing meteorites with lunar surface material we do not want to imply that the surface of the moon has meteoritic composition. But meteorites as well as the lunar surface are exposed to the cosmic radiation, whereas terrestrial material is shielded by the atmosphere. Furthermore, meteorites are much older than terrestrial rocks and in some cases even contain the decay products of short lived isotopes such as I^{129} . In addition, some meteorites hold large amounts of gases with roughly primordial abundance and isotopic composition, again a feature not found in any terrestrial material so far.

In the second part the possible applications of these methods to lunar material will be discussed. It is sometimes necessary to make rather definite assumptions on the history of the lunar surface or certain regions thereof. Choosing a particular model for a working hypothesis does not imply that it is the only one possible. In fact, rare gas measurements might in many cases actually provide a criterion for deciding between different models.

Rather heavy emphasis will be put on the use of the lunar surface as a detector for cosmic ray intensity and spectrum variations of the past. This seems to us a more important problem than some of the more specific applications of rare gas measurements to the history of the lunar surface.

One may ask, why such importance is placed on the study of the rare gases and not on other elements. The reasons are four-fold:

- a.) Rare gas isotopes are produced in various different nuclear transformations and reactions ("short"- and long-lived radioactive decay, spallation reactions, fission).

- b.) There exist 23 stable rare gas isotopes which can be measured all in one sample. A very large mass range in the periodic system is covered by the rare gases.
- c.) The initial abundance of rare gases included in the mineral at the time of its formation is generally very low. Therefore, even the contributions from processes with very low production rates can be measured.
- d.) The experimental methods for the determination of rare gas concentrations and isotopic compositions are the most sensitive of all elements. Amounts as small as 10^5 atoms of xenon may be detected mass spectrometrically (REYNOLDS 1956; CLARK and THODE 1964). The necessary extraction and purification techniques for handling such small gas samples have also been developed. Contamination problems are much less severe than with any other element. Even extended mineral separations can be performed without danger of contamination.

This combination of favorable properties makes the rare gases superior to any other group of elements. Sometimes, their tendency to show diffusion losses may be a certain drawback. But results on meteorites have shown that even for periods of 4.5×10^9 years this is generally no serious problem. Methods for the detection of diffusion losses will be discussed in detail.

2. RARE GAS MEASUREMENTS IN METEORITES.

2.1 INTRODUCTION

In the last few years several review articles have been written on rare gases and age determinations in meteorites and on related subjects (EBERHARDT and GEISS 1960; ARNOLD 1961; ANDERS 1962, 1963, 1964; UREY 1963 a; KIRSTEN, KRANKOWSKY and ZÄHRINGER 1963; REYNOLDS 1963). In this part of our paper we want to outline only the methods and basic results obtained from meteorites. For a more detailed discussion the reader is referred to the above mentioned publications.

In table 1 all known twenty three stable rare gas isotopes are listed. All of them have been found in meteorites. Their amounts may vary from meteorite to meteorite by as much as a factor 100'000. To show the orders of magnitude involved the concentrations in a chondrite and an iron meteorite are listed in table 1. For comparison the concentrations in the terrestrial atmosphere are also given.

Five different sources for rare gases in meteorites have been recognized so far:

- a) Radioactive decay of long lived isotopes in the meteorite
- b) Spallation reactions induced by particle irradiation of the whole meteorite or of meteorite constituents (e. g. chondrules)
- c) Radioactive decay of short lived (extinct) isotopes in the meteorite
- d) Spontaneous or induced fission in the meteorite
- e) Gases trapped in the whole meteorite or in certain fractions.

In the processes a, b, c and d the rare gas isotopes are produced inside the meteorite, or fragments which later accumulated to form the meteorite. The four processes differ in the type of nuclear reaction or

Isotope	Concentration in typical chondrite in 10^{-8} ccSTP/g	Concentration in typical iron meteorite in 10^{-8} ccSTP/g	Concentration in earth's atmosphere in 10^{-8} cc/cc
He ³	33	530	0.0007
He ⁴	1510	1980	524
Ne ²⁰	9.1	6.4	1650
Ne ²¹	9.5	6.3	4.7
Ne ²²	10.5	6.7	160
Ar ³⁶	1.39	21.1	3150
Ar ³⁸	1.16	33.5	588
Ar ⁴⁰	5400	21.8	930000
Kr ⁷⁸	-	-	0.40
Kr ⁸⁰	-	-	2.59
Kr ⁸²	-	-	13.2
Kr ⁸³	~ 0.012	-	13.2
Kr ⁸⁴	~ 0.057	-	64.8
Kr ⁸⁶	~ 0.017	-	19.8
Xe ¹²⁴	0.00014	-	0.0083
Xe ¹²⁶	0.00012	-	0.0077
Xe ¹²⁸	0.0021	-	0.165
Xe ¹²⁹	0.036	-	2.28
Xe ¹³⁰	0.0040	-	0.35
Xe ¹³¹	0.020	-	1.82
Xe ¹³²	0.024	-	2.31
Xe ¹³⁴	0.0094	-	0.90
Xe ¹³⁶	0.0078	-	0.76

Table 1: Stable rare gas isotopes and their concentrations found in a typical chondrite (Richardton), iron meteorite (Charcas) and in the earth's atmosphere. Data from: EBERHARDT and HESS (1960); EBERHARDT and EBERHARDT (1961); GEISS and HESS (1958); REYNOLDS (1960b) and SIGNER and NIER (1961). (Dashes: not measured)

transformation; a and c are basically the same processes, their only difference is the half-life of the radioactive isotope. Thus, a and c data two completely different time intervals and, therefore, it is justified to consider them as two different sources.

Table 2 gives a survey of all stable rare gas isotopes and the processes which have been recognized in meteorites as contributors to these isotopes. In many cases a single isotope may have been produced by several different sources. Since the isotopic composition of a rare gas is characteristic for its source, it is normally possible to decide which or what mixture of the five processes is responsible for the observed gas concentration.

2.2 RADIOACTIVE DECAY OF LONG-LIVED ISOTOPES

2.2.1 General

Only two rare gas isotopes, He^4 and Ar^{40} , are produced in the radioactive decay of a long-lived isotope. (We consider an isotope long-lived if its half-life is longer than a few hundred million years and detectable amounts have thus survived the time interval since the end of nucleosynthetic processes). He^4 is formed in the decay of U^{238} , U^{235} and Th^{232} ; Ar^{40} from the decay of K^{40} . The concentration found in a meteorite, or any other sample, can be expressed by the following equation:

$$C^I = \sum_i c_i \int_{-\infty}^0 v_i \lambda_i p_i(t) \mu_i(t) e^{-\lambda_i t} dt + C_0^I \quad (1)$$

with:

C^I : concentration of daughter product at present time

c_i : concentration of parent isotope today

v_i : number of daughter atoms produced by one decaying parent atom

λ_i : radioactive decay constant

Isotope	Rad. decay long-lived isotope 10 ⁻⁸ cc/g	Spallation 10 ⁻⁸ cc/g	Rad. decay short-lived isotope 10 ⁻⁸ cc/g	Fission 10 ⁻⁸ cc/g	Trapped 10 ⁻⁸ cc/g		
He ³	2000	200			700		
He ⁴		800			2000000		
Ne ²⁰		60			30000		
Ne ²¹		60			100		
Ne ²²		60			3000		
Ar ³⁶		3			650		
Ar ³⁸		3			120		
Ar ⁴⁰	7000				}		
Ar ⁷⁸							
Kr ⁸⁰							
Kr ⁸²							
Kr ⁸³						}	
Kr ⁸⁴							
Kr ⁸⁶							
Kr ¹²⁴							
Xe ¹²⁶		0.00001					
Xe ¹²⁸		0.00002					
Xe ¹²⁹							
Xe ¹³⁰	0.5	}	3.7				
Xe ¹³¹							
Xe ¹³²							
Xe ¹³⁴							
Xe ¹³⁶							
Xe							
							-0.02

Table 2: Recognized sources of stable rare gases in stone meteorites and their observed maximal concentrations.

- $\mu_i(t)$: concentration of parent isotope at time t , relative to today's concentration, corrected for radioactive decay
- $p_i(t)$: probability that daughter atom produced at time t is still in mineral
- C_o^I : concentration of daughter product not due to radioactive decay in situ

Equation (1) holds also for any other daughter product of a long-or short-lived radioactive isotope, e. g. Sr^{87} , Pb^{206} etc. C^I and c_i are the two measurable quantities in a given sample, ν_i and λ_i are known and constant with time. (We assume throughout this paper that the basic coupling constants do not change with time. For a detailed discussion of this subject see PEEBLES and DICKE 1962).

C_o^I , the concentration of the daughter isotope not due to the radioactive decay of the parent in the sample, can sometimes be calculated from the other stable isotopes of the daughter element (e. g. Ar^{40} from Ar^{36} and Ar^{38} ; Sr^{87} from Sr^{84} , Sr^{86} , Sr^{88} ; Pb^{206} , Pb^{207} and Pb^{208} from Pb^{204}). For our further discussion we assume that such a correction has always been applied and that C^I is the concentration due to radioactive decay in situ. $\mu_i(t)$ and $p_i(t)$ are unknown and depend on the origin, and the history of the sample. Generally one has to make specific assumptions on the behavior of the product of these two functions. These have to be inferred from other observations and from certain hypotheses on the origin and life history of the sample in question. Some possible simple models will now be discussed.

2.2.2 The Ideal Case

$$\begin{aligned}\mu_i(t) \times p_i(t) &= 0 & t < T \\ \mu_i(t) = p_i(t) &= 1 & t > T\end{aligned}\tag{2}$$

Under this assumption T can be obtained from the integral of equation (1)

$$C^I = \sum c_i v_i (e^{-\lambda_i T} - 1) \quad * \tag{3}$$

T is called the radiogenic age of the sample, and can always be calculated even if the above mentioned assumptions are not fulfilled. $\mu_i(t)$, $p_i(t)$ or both are zero during the time interval $-\infty < t < T$. This may be due to any one of the following three sample histories:

- a) The sample in question did not yet exist before T and was formed at the time T in its present form. The time interval of formation is short compared to T .
- b) The sample was at such a high temperature before T that all the daughter atoms, formed before T , were lost by diffusion ($p_i(t) = 0$ for $t < T$). At the time T the sample was cooled down rapidly.
- c) The sample was heated at time T to such a high temperature that all the daughter atoms formed before were lost by diffusion. Chemical or mineralogical alterations may or may not have taken place.

For $t > T$ $\mu_i(t)$ and $p_i(t)$ are both equal to one and thus no diffusion losses and no chemical changes should have occurred since the time T . From a single age measurement alone it is not possible to decide if "the ideal case" applies. But the coincidence of two independent ages, such as K-Ar and U/Th-He ages on the same sample, is a good check that the assumptions of the ideal case are fulfilled and the ages are indeed dating one of the three above mentioned events. A disagreement of the two ages

* T is always negative

does not necessarily mean that both are influenced by diffusion loss. Large differences in diffusion coefficients may exist between the two rare gases and thus helium may show diffusion losses whereas argon is not affected. A more detailed discussion will be given below.

2.2.3 The Non-Ideal Case

In the ideal case rather extreme conditions were assumed. Up to a certain time T , the age of the sample, all the daughter atoms produced were lost by diffusion. Afterwards no diffusion loss, or chemical changes occurred. Often, these conditions will not be true. If the grain size and shape distribution, the individual diffusion coefficients of all the mineral phases and their temperature dependence, the parent element distribution in the mineral grains and the temperature history of the sample were known, then the survival probability $p_i(t)$ could be calculated.

For the present discussion we shall assume two different types of diffusion loss histories: continuous loss during the whole time interval since T , or diffusion loss only during the very latest part of the life of the meteorite. We assume a constant concentration for the parent element (constant if corrected for radioactive decay) ($\mu_i(t) = 1$) throughout the time interval $T < t$. This assumption is justified, because the temperatures required for diffusion of the solid parent elements would most likely be so high that any previously contained rare gases would be lost.

2.2.3.1 Continuous Diffusion Loss

By solving the diffusion equation for homogeneous spheres one could calculate the survival probability $p_i(t)$. But, because of the many unknown factors mentioned above, nothing is gained by using the exact solution of the diffusion equation and further mathematical treatment is complicated. Therefore, we assume that a daughter atom has a constant probability to be lost by diffusion in any time interval dt . The survival function will then have the simple form

$$\begin{aligned} p(t) &= e^{-\lambda t} & T < t \\ p(t) &= 0 & t > T \end{aligned} \quad (4)$$

where λ is the diffusion probability. Substituting (4) in equation (1) and assuming only one parent isotope (corresponding to K^{40} - Ar^{40} ages) one obtains for the concentration of the stable daughter:

$$C^I = \nu c \int_T^0 \lambda e^{-(\lambda - \lambda')t} dt \quad (5)$$

and thus

$$C^I = \nu c \frac{\lambda}{\lambda - \lambda'} \left[1 - e^{-(\lambda - \lambda')T} \right] \quad (6)$$

If T_D is the apparent age of the sample, obtained by using equation (3), even if the assumptions of this equation are not fulfilled, then one can easily deduce the following relation between apparent age, true age T and diffusion probability:

$$T_D = \frac{1}{\lambda} \left[\ln\left(1 - \frac{\lambda'}{\lambda}\right) - \ln\left(e^{-(\lambda - \lambda')T} - \frac{\lambda'}{\lambda}\right) \right] \quad (7)$$

For $T \rightarrow -\infty$; T_D becomes

$$T_D = \frac{1}{\lambda} \ln \left(1 - \frac{\lambda}{\mathcal{K}} \right) \quad \text{if } \mathcal{K} > \lambda \quad (8)$$

Thus, the apparent age T_D of a sample is finite even if the true age would be infinite (if $\mathcal{K} > \lambda$). This simple model gives a satisfactory agreement with exact diffusion theory if $\mathcal{K} = 16 D/a^2$ is assumed. (D: diffusion coefficient, a: radius of grain). Figure 1 shows the true age/apparent age relation calculated with this approximation. For comparison the true age/apparent age relation calculated from exact diffusion theory is also given (GOLES, FISH and ANDERS 1960). The agreement is very good for all practical purposes.

The advantage of this analytical treatment is that it can easily be applied to any grain size distribution. The above derived relations assumed uniform grain size, which is certainly not true in any real sample. If $N(a) da$ is the differential grain size distribution, then the concentration of a stable daughter isotope in a sample will be (c independent of a):

$$C^I = VC \frac{\int_0^\infty a^3 N(a) \frac{\lambda}{\lambda - \mathcal{K}} (1 - e^{(\mathcal{K} - \lambda)T}) da}{\int_0^\infty a^3 N(a) da} \quad (9)$$

Very little is known at the present time about the grain size distributions in meteorites and thus a further evaluation of this equation is not possible.

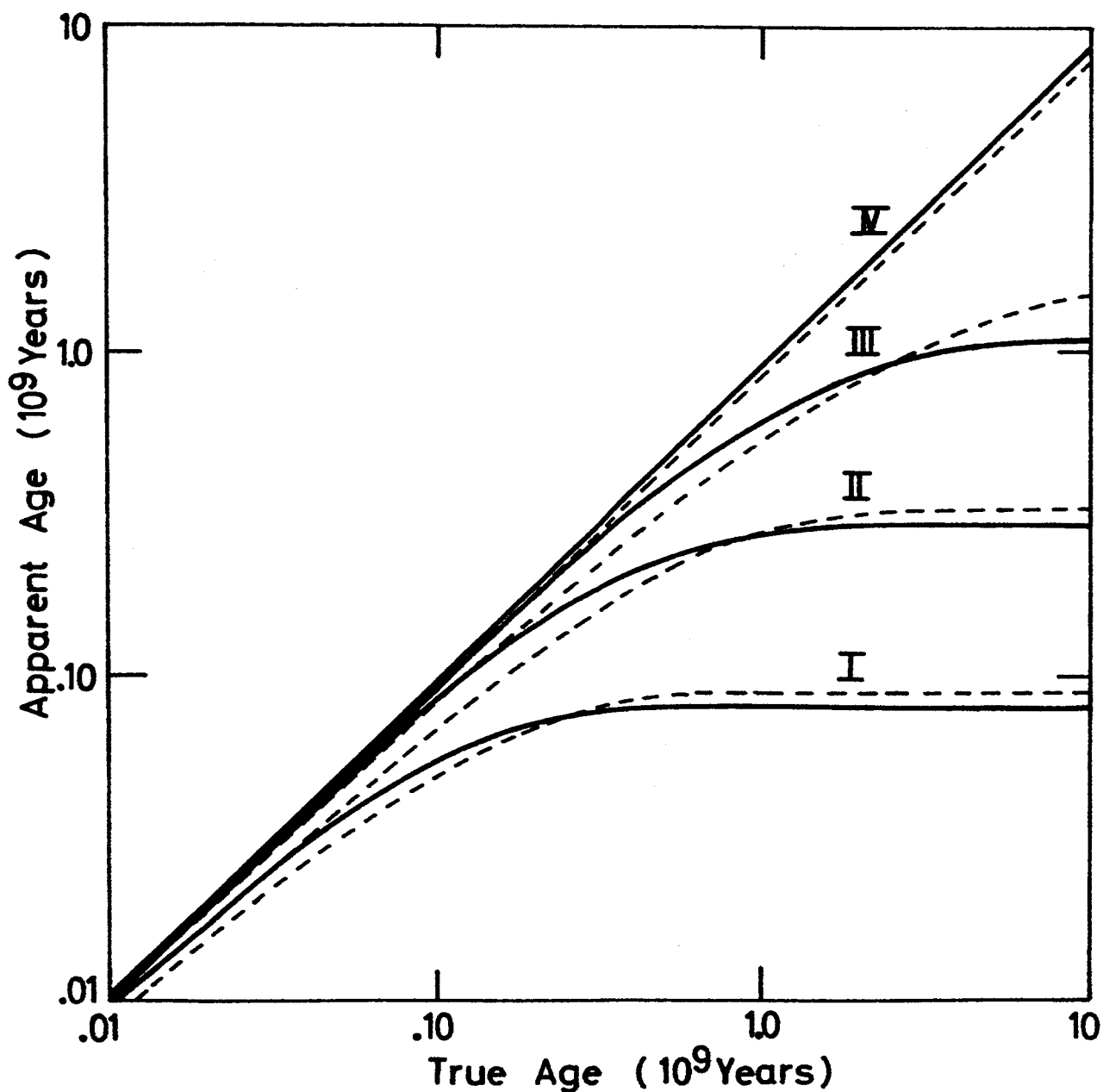


Figure 1. True age / apparent age relation for continuous diffusion loss from spherical grains. Solid lines calculated with approximation given in text (equation (7)), dashed curves with exact diffusion theorie (GOLES, FISH and ANDERS 1960). D/a^2 -values for the different curves are I : $2.5 \times 10^{-17} \text{ sec}^{-1}$; II : $7.5 \times 10^{-18} \text{ sec}^{-1}$; III : $2.5 \times 10^{-18} \text{ sec}^{-1}$; IV : $2.5 \times 10^{-19} \text{ sec}^{-1}$.

2.2.3.2 Recent Diffusion Loss

It is known that stone meteorites were broken out of a larger body very late in their history (only several million years ago) (see 2.2.4). During this break up moderate heating may have taken place and, therefore, it seems reasonable to discuss a model where [diffusion loss of radiogenic rare gases] occurred very late in the meteorite history. Thus we would have

$$\begin{aligned} p(t) &= 0 & t < T \\ p(t) &= p & T < t < \infty \end{aligned} \quad (10)$$

and the concentration will be

$$C^I = \nu c p (e^{-\lambda T} - 1) \quad (11)$$

In a corresponding manner one obtains for the apparent age T_D

$$T_D = -\frac{1}{\lambda} \ln (1 - p + p e^{-\lambda T}) \quad (12)$$

p can be calculated from the exact diffusion theory for homogeneous spherical grains and turns out to be (CARSLAW and JAEGER 1959):

for small diffusion loss:

$$p \approx 1 - \frac{6}{\pi^{1/2}} \left(\frac{D\tau}{a^2} \right)^{1/2} + 3 \frac{D\tau}{a^2} \quad 0.15 \leq p \leq 1 \quad (13)$$

for large diffusion loss:

$$p = \frac{6}{\pi^{1/2}} \exp\left(-\tau^2 \frac{D\tau}{a^2}\right) \quad p < 0.15 \quad (14)$$

is the time interval during which the diffusion loss has taken place and according to our assumptions $|\tau| \ll |T|$.

From equations (6), (11) and (14) it can be easily seen that the concentration C^I is very strongly size dependent, decreasing extremely rapidly with decreasing grain size. Figure 2 shows the calculated grain size dependence for continuous and for recent diffusion loss. This strong grain size dependence should be very useful for checking a sample for diffusion loss. If the assumptions of the ideal case hold then the age should not be grain size dependent. Thus, measuring the age of at least two grain size fractions will indicate if diffusion loss has occurred. When a grain size/age curve of a sample has been experimentally determined it should be possible to calculate the true age by extrapolating to large grain sizes. Because of cross contamination of finer grain size fractions by fragments of larger grains it will probably not be feasible to use the difference of the two curves for deciding between the two diffusion histories.

Ages of different minerals and constituents (e.g. chondrules and matrix) could also be determined and their agreement used as a test for diffusion losses. This method implies that the true ages of different minerals and constituents do agree, an assumption which is not necessarily true. Different grain sizes of the same constituent are more likely to have the same true age and thus the grain size measurement should be the better indicator for diffusion losses.

2.2.4 Results

K-Ar and U/Th-He ages have been measured on about 70 meteorites. The main results obtained can be summarized as follows:

- 1.) Most of the K-Ar-ages of chondrites lie between 3.5 and 4.5×10^9 y (GEISS and HESS 1958; KIRSTEN, KRANKOWSKY and ZAEHRINGER 1963).
- 2.) U/Th-He-ages are either concordant to or lower than K-Ar-ages, in the latter case indicating diffusion loss (EBERHARDT and HESS 1960; ANDERS 1962).

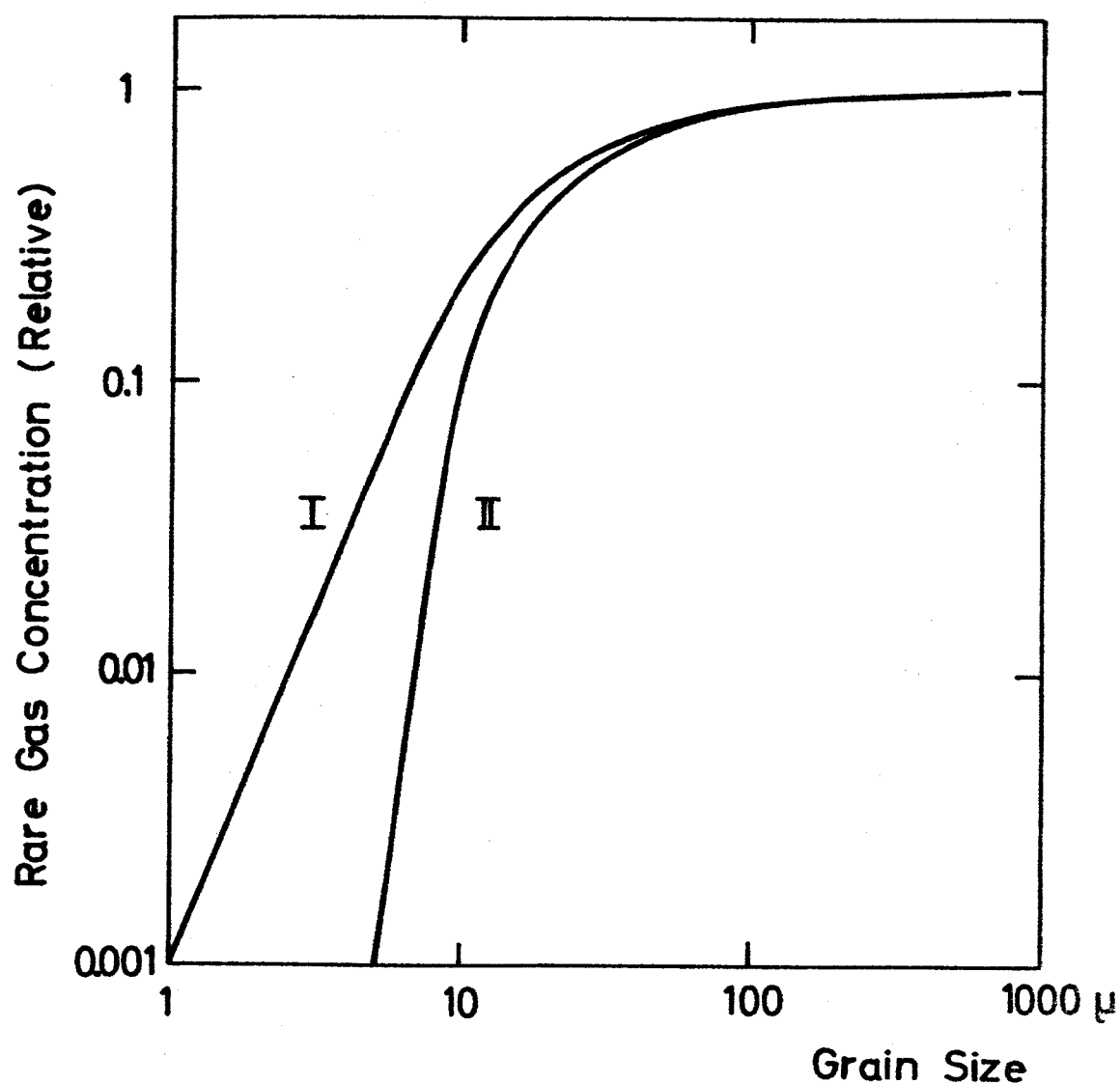


Figure 2. Dependence of diffusion loss on grain size. Curve I for continuous diffusion loss ($D = 10^{-24} \text{ cm}^2 \text{ sec}^{-1}$, true age $4.5 \times 10^9 \text{ y}$), curve II for recent diffusion loss ($D\tau = 1.8 \times 10^{-7} \text{ cm}^2$, true age $4.5 \times 10^9 \text{ y}$).

- 3.) The age distribution of the various classes of chondritic meteorites seems to be different, implying different histories (EBERHARDT and GEISS 1964 a; ANDERS 1964).
- 4.) A few meteorites with low concordant Ar and He ages exist, indicating a special event in the history of the meteorite parent bodies (ANDERS 1964).

2.3 SPALLATION PRODUCED RARE GASES

2.3.1 General

During the time a meteorite is travelling in interplanetary space as a small body it is exposed to the particle radiation. Nuclear reactions, mainly spallation reactions induced by the higher energetic cosmic radiation (above several ten MeV) lead to the formation of new radioactive and stable isotopes. The direct production rate of a stable or a radioactive isotope can be calculated from:

$$P^I(\vec{r}, t) = \sum_i \sum_k \int_0^\infty \sum_i^I(E) f_k(\vec{r}, E, t) dE \quad (15)$$

where:

$P^I(\vec{r}, t)$: Production rate of isotope I at position r and time t

$\sum_k \sum_i^I(E)$: Macroscopic production cross section of isotope I from element i by particle k of energy E .

$f_k^I(\vec{r}, E, t)$: Time dependent energy spectrum of particle k at position r of the sample.

The index i is to be summed over all elements in the target and the index k over all types of particles present at the location of the sample (protons, neutrons, deuterons, α -particles, mesons etc.).

For the calculation of the concentration of spallation produced isotopes the indirect production by the radioactive decay of other spallation isotopes will also have to be taken into account (if not already included in $\sum_i^I(E)$). Thus, the total production rate of a stable isotope, such as Ne^{21} or Ar^{38} , will be the sum of all the direct production rates for all the elements on the corresponding isobar.

The irradiation spectrum ${}_k^f(\vec{r}, E, t)$ at the sample position could be calculated from the spectrum ${}_k^F(E, t)$ of the primary cosmic radiation if the shielding geometry were known:

$${}_k^f(\vec{r}, E, t) = {}_k^S(\vec{r}) {}_k^F(E, t) \quad (16)$$

or

$${}_k^F(E, t) = \overline{{}_k^S(\vec{r})} {}_k^f(\vec{r}, E, t) \quad (17)$$

where ${}_k^S(\vec{r})$ and $\overline{{}_k^S(\vec{r})}$ are the shielding factor operators, transforming the primary spectrum into the actual spectrum at the sample position and vice versa. The calculation of production rates according to equation (15) and (16) is extremely difficult and tedious (ARNOLD, HONDA and LAL 1961). Furthermore, the necessary excitation functions and other data are in many cases not yet known.

Simpler models have been used to avoid these difficult calculations. Instead of a continuous energy spectrum only two discrete energies were assumed representing the high energy primary radiation and the low energy secondary particles, including pions, produced in the spallation itself (EBERT and WAENKE 1957; HOFFMAN and NIER 1958; SIGNER and NIER 1960; GOEL 1962). No further distinction is made between the different particles such as nucleons and mesons. A depth dependence of the production rate along a parallel beam of the form

$$P_{(x)}^I = A_1 e^{-\beta_1 x} + x^\alpha A_2 e^{-\beta_2 x} \quad (18)$$

is in good agreement with the observed radial distribution of spallation products in iron meteorites. A_1 , A_2 , β_1 , β_2 and α are constants depending on the chemical composition of the target material and the isotope I. An exponent $\alpha > 0$ takes into account a strong contribution by secondaries including pions. A good agreement for the He^3 , He^4 , Ne^{20} and Ar^{38} depth dependence was obtained with $\alpha = 0$ (SIGNER and NIER 1960).

Using the depth dependence as given in equation (18) it is possible to calculate the production rate in a meteorite sample. Important for our discussion of spallation effects on the lunar surface will be the semi infinite planebounded slab. Integrating equation (18) for an omni-directional cosmic ray flux leads to (for $\alpha = 0$):

$$P^I(x) = A_1^* \left(e^{-\beta_1 x} - \beta_1 x \int_1^\infty \frac{e^{-\beta_1 x \xi}}{\xi} d\xi \right) + A_2^* \left(e^{-\beta_2 x} - \beta_2 x \int_1^\infty \frac{e^{-\beta_2 x \xi}}{\xi} d\xi \right) \quad (19)$$

This form is not very well suited for further analytical treatment. Without introducing an appreciable error this expression can be approximated by:

$$P^I(x, t) = P^I(0, t) \left[B_1 e^{-\mu_1 x} + B_2 e^{-\mu_2 x} \right] \quad (20)$$

The agreement of this approximation with equation (19) is very good, except at the immediate surface and at great depth. However, the deviation at the surface is relatively small and for great depth the absolute production rates are small. Furthermore, equation (18) is also only an approximation and no experimental verification is available in these regions.

The concentration of a radioactive and stable isotope in a sample can be obtained from the production rate $P^I(x, t)$:

for stable isotopes:

$$C^{I_s}(x) = \int_{T_R}^0 P^{I_s}(x, t) dt \quad (21)$$

for radioactive isotopes:

$$C^{I_r}(x) = \int_{T_R}^0 P^{I_r}(x,t) e^{\lambda t} dt \quad (22)$$

T_R is the time at which the irradiation of the sample commenced. If we assume that at this time the irradiation of the sample was started by the sudden removal of excessive shielding and that no changes in the production rate occurred afterwards the "radiation age" T_R of the sample can be calculated from:

$$T_R = - \frac{C^{I_s}(x)}{P^{I_s}(x)} \quad (23)$$

The production rate $P^{I_s}(x)$ can be obtained from the activity, $\lambda C^{I_r}(x)$, of a radioactive isotope in the sample and from the ratio of the relative production cross sections \sum_s and \sum_r

$$P^{I_s}(x) = \lambda C^{I_r}(x) \frac{\sum_s}{\sum_r} \quad (24)$$

and thus:

$$T_R = - \frac{C^{I_s}(x)}{\lambda C^{I_r}(x)} \frac{\sum_s}{\sum_r} \quad \text{if } \lambda T_R \ll -1 \quad (25)$$

The ratio of the production cross sections is not very energy dependent and can easily be inferred from target measurements if genetically related pairs such as H^3-He^3 , $Cl^{36}-Ar^{36}$ are used, where a large amount of the stable isotope is produced via the radioactive isotope (about 80 per cent for Ar^{36}).

2.3.2 Results

Radiation ages have been determined on a large number of meteorites. The important results obtained are the following:

- 1.) Radiation ages of all meteorites investigated so far are much lower than the radioactive ages, indicating that the meteorite has spent most of its life-time embedded in a larger body (BEGEMANN, GEISS and HESS 1957; GEISS 1957; EBERHARDT and HESS 1960; HINTENBERGER, KOENIG and WAENKE 1962; KIRSTEN, KRANKOWSKY and ZAEHRINGER 1963).
- 2.) Radiation ages of iron meteorites are generally an order of magnitude higher than those of stone meteorites (UREY 1959).
- 3.) Radiation ages for both stones and irons show a continuous and extended distribution (EBERHARDT and HESS 1960; KIRSTEN, KRANKOWSKY and ZAEHRINGER 1963).
- 4.) Radiation ages of the low-iron chondrites (hypersthene chondrites) show a clustering around 20 m. y., indicating a distinct break up process of a body of at least 5 km in diameter (GEISS, OESCHGER and SIGNER 1960; EBERHARDT and GEISS 1964).
- 5.) Radiation ages of the aubrites as a group are different from those of the other classes of stone meteorites. Thus, their origin and history must be different (EBERHARDT, EUGSTER and GEISS 1964 b).
- 6.) Radiation ages of the various iron meteorite classes seem to be different, the hexahedrites showing generally lower ages (VILCSEK and WAENKE 1961; ANDERS 1962).

2.4 SHORT LIVED ISOTOPES

The isotope I^{129} has a half-life of 17 m. y. and decays to Xe^{129} . Thus, if the interval between the end of nucleosynthetic processes and the formation of the meteorite or its minerals was not longer than a few half-lives, some I^{129} might have been incorporated into the meteorite together with the stable I^{127} . Its decay would lead to an excess of Xe^{129} in the meteorite.

REYNOLDS (1960) was indeed able to show that the chondrite Richardson contains an excess of Xe^{129} . Since then many more meteorites have been found with such an excess (REYNOLDS 1960 a; ZAEHRINGER 1961; JEFFERY and REYNOLDS 1961; MERRIHUE, PEPIN and REYNOLDS 1962; CLARKE and THODE 1964). Making specific assumptions on the type of nucleosynthetic processes responsible for the I^{129} , the time interval between the end of such processes and the formation of the meteorite (beginning of xenon retention) can be calculated from the ratio $Xe^{129}_{\text{excess}}/I^{127}$. These intervals lie between 60 m. y. and 250 m. y.

Several other isotopes with similar half-lives exist. No effects could be found for Tl^{205} (ANDERS and STEVENS 1960). The reported anomalies in the isotopic composition of Ag^{107} (MURTHY 1960, 1962) are probably due to fractionation effects in the mass spectrometer (CHAKRABURTTY, STEVENS, RUSHING and ANDERS 1964). In both cases the primordial concentration of the daughter element in the meteorite is much higher than that of Xe^{129} . Pu^{244} and some other transuranian elements will be discussed in the next chapter.

2.5 FISSION PRODUCED ISOTOPES

The heavy isotopes of xenon in meteorites (Xe^{136} , Xe^{134} , Xe^{132} , Xe^{131}) show a different isotopic composition than terrestrial xenon (REYNOLDS 1960 b, 1963; ZAEHRINGER 1961; MERRIHUE, PEPIN and REYNOLDS 1962; MANUEL and KURODA 1964; REYNOLDS and TURNER 1964). The earth's atmosphere is enriched in these isotopes relative to meteorites. The anomalies are most likely due to the spontaneous fission of Pu^{244} or some other long lived transuranium elements (KURODA 1960). The contribution of these processes to the terrestrial atmosphere with its high depletion factor is much higher than for the meteorites. Photofission and perhaps neutron induced fission may also have played an important role.

At the present, all these phenomena are not yet fully understood, in particular also the anomalies of the light xenon isotopes (Xe^{124} , Xe^{126}). But they have already given important clues on the early history of our solar system and the processes which have contributed to the formation of certain elements.

2.6 TRAPPED GASES

GERLING and LEVSKII (1956) were the first to discover a meteorite (Pesyanoe) containing large quantities of He^4 , Ne^{20} , Ne^{22} , Ar^{36} and Ar^{38} . From the absolute amounts and the isotopic composition they concluded that these excessive amounts of light rare gases could not have been produced by any of the previously mentioned four processes, but were probably gases dissolved in the minerals of the meteorite during its formation. Since then about ten additional meteorites have been found with high contents of light rare gases (ZAEHRINGER and GENTNER 1960; KOENIG, KEIL, HINTENBERGER, WLOTZKA and BEGEMANN 1961; KOENIG, KEIL and HINTENBERGER 1962; MANUEL and KURODA 1964; EBERHARDT and GEISS 1964 c), and about fifteen others with high concentrations of trapped heavy rare gases (argon, krypton and xenon) (REYNOLDS

1960 b, 1960 c; STAUFFER 1961; ZAEHRINGER 1962 a; SIGNER and SUESS 1963; EBERHARDT and GEISS 1964 a).

Some typical examples are listed in table 3. The difference between the two previously mentioned groups of meteorites containing trapped rare gases is rather obvious. Whereas Tysnes Island and Khor Temiki contain large amounts of trapped He^4 , Ne^{20} and Ne^{22} , practically none can be detected within the limits of error in Mezö Maderas and Tieschitz. All four contain trapped argon but only the latter two large amounts of excessive krypton. The distinction between the two classes is not always as obvious as in the examples listed in table 3, and mixtures of both components may occur. SIGNER and SUESS (1963) were the first to recognize these two components and accordingly we shall also call the light component solar and the heavy planetary. A justification for this nomenclature will be given later.

For a more detailed discussion of trapped gases and their origin it is useful to consider their relative and isotopic abundances and not their absolute concentrations. The latter depend on secondary parameters and may not be representative of the mother reservoir of the trapped gases. The relative and isotopic abundance, on the other hand, is directly related to the origin and trapping mechanism. Figure 3 shows the abundance of trapped He^4 , Ne^{20} and Kr relative to Ar^{36} . Only selected analyses have been plotted to avoid cluttering. The meteorites containing large amounts of the solar component show abundances similar to the cosmic ones, which, in turn, are derived from solar and stellar abundances (ALLER 1961). Khor Temiki agrees within a factor of two, whereas the other meteorites show larger deviations. It is obvious from figure 3 that the deviations are mass dependent, the depletion being strongest for helium, less for neon and showing an enrichment of krypton relative to argon. Thus, the solar component must have been derived from a reservoir with nearly cosmic abundances, which was never strongly fractionated. It even seems possible to assume that the solar component has cosmic abundances and the observed slight fractionation is due to diffusion losses after the gas has been trapped in the meteorite.

Meteorite	^3He	^4He	Ne^{20}	Ne^{21}	Ne^{22}	Ar^{36}	Ar^{38}	Ar^{40}	Kr
Tysnes Island	39	112000	413	3.2	38.6	25.5	5.2	4800	<0.1
Khor Temiki	107	49500	130	22.4	34.2	3.8	1.4	3950	<0.1
Mezö Maderas	53	860	11.8	11.1	12.6	55	12	1250	~0.5
Tieschitz	47	1700	9.0	7.5	9.1	23	5.3	2300	~0.3

Table 3: Typical examples of meteorites with trapped gases. The first two belong to the group containing trapped light rare gases, the last two to the one containing heavy rare gases. Preliminary data from EBERHARDT and GEISS (1964c) (for final data see mentioned publication).

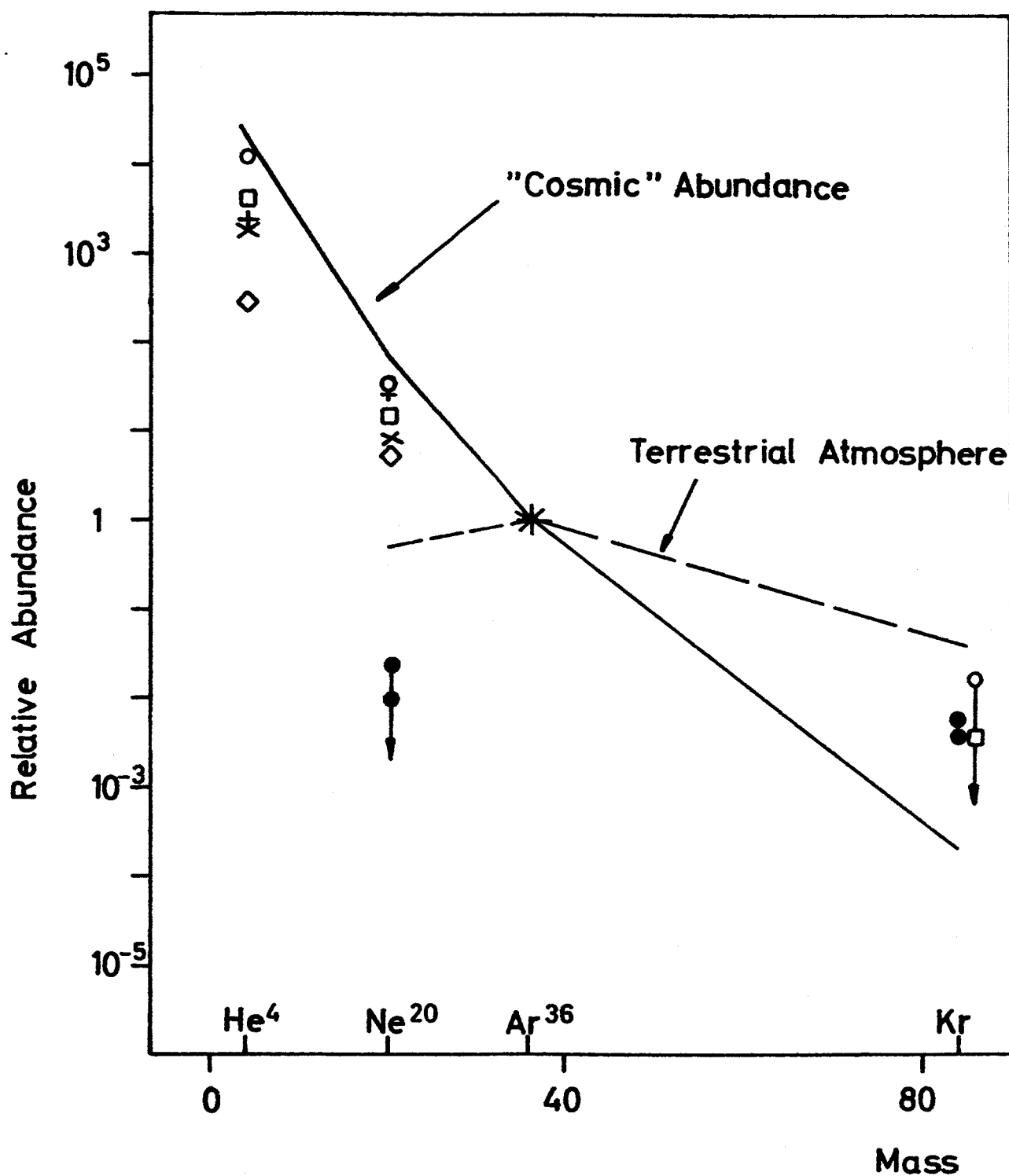


Figure 3. Concentration of trapped rare gases relative to trapped Ar^{36} . Open symbols represent meteorites with trapped light rare gases, solid symbols meteorites with trapped heavy rare gases. Arrows indicate, that values are only upper limits. (Preliminary data from EBERHARDT and GEISS 1964c).

The planetary component shows an abundance pattern similar to the terrestrial atmosphere. Therefore, the corresponding reservoir must have been heavily fractionated before the gases were trapped in the meteorite. (We assume that the trapping mechanism itself is not responsible for any strong fractionation).

For a further discussion of the origin of these trapped gases the trapping mechanism itself should be known. Not much experimental evidence is available and it is not known if the trapping mechanism is the same for both components. The planetary component seems to be present in all meteorites belonging to certain meteorite classes (carbonaceous chondrites and enstatite chondrites). Many meteorites containing the solar component have a dark-light structure, only the dark phase holding trapped gases. These meteorites also show signs of strong shock waves. FREDRIKSSON, DE CARLI, PEPIN, REYNOLDS and TURNER (1964) have shown that shock waves may trap rare gases in meteorites, but it has not yet been proven that the trapped gases found in meteorites were indeed incorporated in the minerals by this mechanism.

Recently it has been shown that the concentration of trapped gases in an enstatite achondrite seems to increase with decreasing grain size. (EBERHARDT, GEISS and GROEGLER 1964). This would indicate, that the trapped gases are concentrated on the surface of the individual mineral grains. It is not known if shock implantation of the rare gases would result in such a concentration on the surface. An alternative trapping mechanism leading to such a surface effect is the bombardment of the individual grains with low energy rare gas atoms or ions (several hundred eV to MeV). The solar wind would be an ideal source for such low energy particles, and was certainly present throughout the whole history of the solar system with the possible exception of its earliest stage. The mineral grains must have been exposed to the solar wind either as free particles in interplanetary space or as immediate surface layer of a body without an atmosphere and strong magnetic field (e.g. an asteroid or a moon).

An irradiation time of less than a year would be sufficient to account for the observed concentrations, assuming present day fluxes (quiet sun).

Afterwards, the grains must have accumulated and been compacted to form the present day meteorite. It could be that shock waves from impacts or collisions might be responsible for this compacting.

Nothing yet is known on the trapping mechanism and the origin of the planetary component.

3. POSSIBLE APPLICATIONS TO THE

LUNAR SURFACE

3.1 RADIOACTIVE AGE DETERMINATIONS

3.1.1 Age of Surface Features

Radioactive age determinations can be used to date important events in the lunar history. From visual observations SHOEMAKER and HACKMAN (1960) have already established a stratigraphy of certain lunar regions. They were able to recognize a sequence of lunar geological periods (see table 4). Each period is characterized and named after an important surface feature. Other lunar features can be ranged in this stratigraphic time scale by careful comparative analyses of their position in the lunar surface strata.

This time scale is of course relative and nothing can be said about the absolute age of any time period. Several attempts have been made to deduce relative and absolute ages from crater frequencies (OEPIK 1960; KREITER 1960; SHOEMAKER, HACKMAN and EGGLETON 1961). Absolute dating by the crater frequency method depends on many poorly known factors, such as the present day meteorite influx and its variations in the past, the impact mass/crater size relation, crater erosion rates on the lunar surface, etc. Especially the meteorite influx in the past is completely unknown and recent results obtained on meteorites (EBERHARDT and GEISS 1964 a; ANDERS 1964) make it very doubtful if the influx has been at all constant, or if it can be approximated by a smooth function. The ages deduced from crater frequency statistics may, therefore, have at the present rather large errors.

Copernican System
Eratosthenian System
Procellarian System
Imbrian System
Archimedian Series
Apenninian Series
Pre-Imbrian System
Ptolemaic Series
Highlandian Series

Table 4: Lunar stratigraphic column.
From DODD, SALISBURY
and SMALLEY (1963). (Pre-
sent at top).

It is, thus, very important to determine the absolute age of lunar surface features by one or several of the radioactive dating methods. If the absolute ages of some of the large craters and maria, defining the stratigraphic lunar time scale were known, it would be possible to obtain the approximate absolute age of other features by their position in the stratigraphic time scale.

Absolute dating requires suitable material with a history corresponding to the "ideal case" discussed in chapter 2.2.2. With the evidence at present available it is rather difficult to decide whether the maria contain any such material. But absolute dating of impact craters will certainly be feasible. The impact generates strong shock waves resulting in heating and even complete melting of some of the surrounding rocks. If the temperature is high enough all the previously accumulated rare gases, in particular He^4 and Ar^{40} , will be lost. Thus, the condition $p_i(t) = 0$ for t smaller than the age of the crater holds. If the heated material has a suitable structure and has never been heated again, it will retain all rare gases formed after the impact. GENTNER, LIPPOLT and SCHAEFFER (1963) and GENTNER, LIPPOLT and MUELLER (1964) have shown that it is indeed possible to date terrestrial impact craters with the K-Ar method, using the glassy material formed by the impact. In favorable cases less than 0.1 per cent of the previously contained Ar^{40} remained in the glass after the impact.

Absolute dating of the impact craters should thus be no problem, if suitable material is collected. The age of maria or other lunar features could, of course, be derived from the age of the superimposed impact craters. A mare must be older than the oldest crater it contains and younger than the youngest crater it completely covers. A drawback of this method is the large number of analyses it may require.

If enough lunar craters are dated it will be possible to establish an absolute age/crater frequency relation and thus calculate the meteorite

influx in the past. This knowledge will be important for our understanding of the history of the solar system. We would like to point out that present day meteorite statistics and even terrestrial impact crater statistics will never be able to provide this information. Meteorites, especially the stone meteorites, have a rather short life expectancy (see 2.3.2). Therefore, the meteorite influx at any time is probably governed by the infrequent breakup of larger bodies and may thus show erratic variations during the life of the solar system. Terrestrial impact crater statistics, for craters older than a few hundred million years, will be heavily biased by erosion and geological activity.

When the history of the cosmic radiation in the past has once been established (see 3.2) it will be possible to use also spallation produced isotopes for the dating of the lunar surface. This method may especially be important for relatively dating two events, such as the age difference of two overlapping craters. A greater accuracy may be obtained for events far in the past than with direct absolute dating. To be suitable for this dating method, material with the following characteristics must be available:

- 1.) The event to be dated must suddenly expose the material to the cosmic radiation by removing any excessive shielding.
- 2.) The remaining shielding should be constant with time or its variation should be known.
- 3.) The material should not contain any spallation rare gases at the moment of exposure (no early irradiation).

By suitably interpreting equation (35), (known cosmic ray flux, unknown irradiation time), the age of the material can be calculated. A more detailed discussion of cosmic ray effects and of the availability of suitable material will be given below.

3. 1. 2 Age of the Moon

A determination of the age of the moon requires that a certain amount of differentiation has taken place during its formation or shortly thereafter. Studies of the uranium-lead and the rubidium-strontium system might then lead to a determination of the age of the moon, in a similar manner as to the determination of the age of the earth (HOLMES 1946, 1949; HOUTERMANS 1946, 1947, 1953; VINOGRADOV, ZADOROZHNYI and ZYKOV 1952; PATTERSON, TILTON and INGRAM 1955; PATTERSON 1956; RUSSELL and ALLAN 1956; EBERHARDT and GEISS 1960; GAST 1960, 1962). Of course, also the methods involving rare gases will be useful for inferring the age of the moon.

Radioactive dating of lunar surface rocks and their mineralogical and morphological constituents will yield their absolute ages. Assuming ideal conditions this will correspond to the time of their formation or their last heating. It is not, a priori, certain that their formation or last heating really has occurred on the moon itself. Today it is generally assumed that the lunar surface was heavily bombarded with smaller objects during the earlier history of the solar system (UREY 1962 b). These objects may be older than the moon. Depending on the impact conditions they may or may not have lost their previously accumulated radiogenic gases. If the moon had then still an atmosphere, even quite large objects might have survived the impact. Depending on surface conditions (thick dust layer) smaller particles might survive even in the absence of an atmosphere. Ice surrounding individual particles might act as "shock absorber". Thus samples older than the moon might be found on the lunar surface, and additional evidence, mineralogical, geological etc. will be required for deciding what event is actually dated.

The time interval between the end of the nucleosynthetic processes responsible for the presence of I^{129} and the formation and cooling of the minerals in question might be obtained with the Xe^{129} method. Fission

produced heavy xenon isotopes will give similar information. A priori, it is again impossible to decide whether this will date a lunar event or the more general accumulation of small bodies or mineral fragments in the solar system which later formed the lunar surface.

3.2 THE HISTORY OF THE PARTICLE RADIATION FIELD IN THE SOLAR SYSTEM

3.2.1 General

Our knowledge of the history of the cosmic radiation has been derived to a large extent from measurements on meteorites. But meteorites are by no means ideal targets. Their irradiation history is not known a priori and has to be deduced from the spallation isotope measurements themselves. No meteorites are known with radiation ages much higher than 10^9 y and thus no information can be obtained covering most of the lifetime of the solar system. A much more suitable target for recording the history of the cosmic radiation is the lunar surface. The formation of craters by the impact of meteorites and also by ejected material from the larger lunar craters, exposes fresh material to the cosmic radiation at a known time (age of the crater). At the same time material which has been exposed is suddenly shielded. These processes were certainly going on during the whole life of the moon and thus most likely will cover the whole period from about 4.5×10^9 y to the present. A certain complication of this simple radiation history may be given by a slow turnover and erosion of the surface. For the moment we will neglect this effect and discuss its influence later (see 3.3).

3.2.2 Intensity Variations

We shall now discuss three different methods which might be useful for detecting intensity variations of the cosmic radiation in the past.

3.2.2.1 Single Crater Method

Let us assume that a single crater is formed at time T by an impact on a surface with a homogeneous irradiation history. Fig. 4 shows a schematic cross section through the crater. From the point of view of cosmic ray induced effects we have to distinguish three different classes of material:

- a.) Material not affected by crater formation. This is material far away from the crater, which is not covered by more than a few centimeters of ejecta.
- β.) Material covered by a thick layer of ejecta, and thus shielded from the cosmic radiation.
- γ.) Freshly exposed material which was not irradiated before the crater was formed.

The following equations for the concentration of a stable spallation product in a sample at irradiation depth x hold in these three regions:

$$\text{region } \alpha \quad C_{\alpha}^{I_s}(x) = \int_{-\infty}^0 P^{I_s}(x,t) dt \quad (26)$$

$$\text{region } \beta \quad C_{\beta}^{I_s}(x) = \int_{-\infty}^T P^{I_s}(x,t) dt \quad (27)$$

$$\text{region } \gamma \quad C_{\gamma}^{I_s}(x) = \int_T^0 P^{I_s}(x,t) dt \quad (28)$$

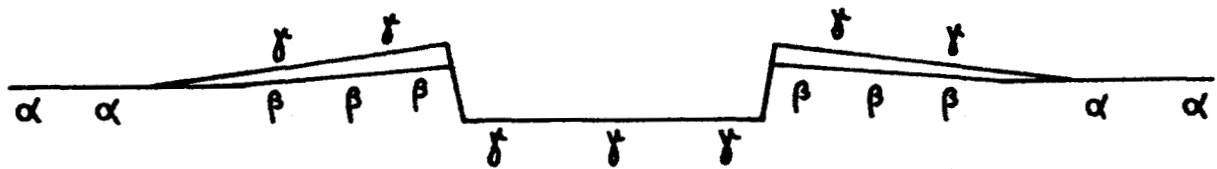


Figure 4. Schematic cross-section through single crater (not to scale). For definition of regions α , β and γ see 3.2.2.1.

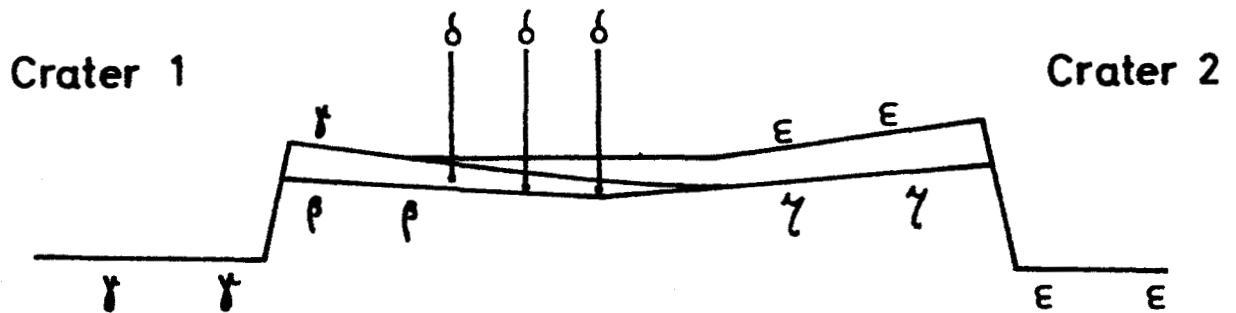


Figure 5. Schematic cross-section through two crater-system (not to scale). For definition of regions α , β , γ , δ , ϵ and ζ see 3.2.2.2.

The concentration C^{Is}_y (y) in a depth y can easily be calculated using equation (20), and thus the samples do not necessarily have to come from the same depth. From equations (26), (27) and (28) it can be seen that

$$C^{Is}_x(x) = C^{Is}_\beta(x) + C^{Is}_\gamma(x) \quad (29)$$

independent of cosmic ray intensity and spectrum. This relation gives an experimental check for the above made assumptions and also for diffusion losses. The age T of the crater can be determined by glass dating using the K-Ar method (see 3.1). When an absolute stratigraphic lunar time scale has been established, T could also be obtained from the stratigraphy of the crater.

Today's production rate $P^I(x, 0)$ can easily be measured from the concentration of a radioactive spallation isotope, e.g. H^3 or Cl^{36} . Also, if the chemical composition and the position of the sample relative to the surface are known $P^I(x, 0)$ can be calculated according to equation (20). If the cosmic ray intensity and spectrum has not changed since T, then

$$P^{Is}(x, 0) = P^{Is}(x, t) \quad (30)$$

and thus

$$C^{Is}_y(x) = T \times P^{Is}(x, 0) \quad (31)$$

In this equation all three variables can be determined experimentally and thus the validity of our assumption of a constant cosmic ray intensity checked. If the intensity has not been constant then the average production rate, $\overline{P^{Is}(x, T)}$, during the time interval $T < t < 0$ can be calculated:

$$\overline{P^{Is}(x, T)} = \frac{C^{Is}_y(x)}{T} \quad (32)$$

Assuming that the spectrum of the cosmic radiation did not vary with time, then we can separate the spectrum into a time dependent and an energy dependent part:

$$F(E,t) = \phi(t) g(E) \quad \phi(0) = 1 \quad (33)$$

Then the production rate of an isotope at time t was

$$P^I(x,t) = P^I(x,0) \phi(t) \quad (34)$$

and

$$\overline{P^{Is}(x,t)} = \frac{P^{Is}(x,0)}{T} \int_0^T \phi(t) dt \quad (35)$$

If similar measurements are performed on a number of craters with different ages T the average production rate, $\overline{P^{Is}(x, T)}$, of an isotope will be known experimentally as a function of the exposure time. Differentiating equation (35) one obtains

$$\phi(t) = - \frac{1}{P^{Is}(x,0)} \frac{d}{dT} \left[T \overline{P^{Is}(x, T)} \right] \quad (36)$$

Thus the cosmic ray intensity at any time in the past could be calculated, the only unknown in the above equation being $\phi(T)$. This method is suitable for the detection of slow variations, but not for short bursts or oscillations (see 3.2.2.4).

3.2.2.2 Two Crater Method

The single crater method discussed above is an integral method, comparing the average production rates over different time intervals. The drawback of this method is that these time intervals are always leading to the present and thus this method is not very sensitive for detecting cosmic ray variations far in the past. Much more suitable for this purpose are measurements on two overlapping craters. Such craters are not infrequent, even large ones exist with clearly overlapping ejecta blankets (e. g. Aristillus and Autolycus; Ptolemaeus and Alphonsus, Aristoteles and Eudoxus etc.). Certainly a very great number of overlapping smaller craters can be found which are still large enough to be useful for such studies.

Figure 5 shows a schematic cross section through a system of two craters. We assume that the craters are so large that the ejecta blankets are adequate for completely shielding the cosmic radiation. In regard to irradiation history we have to distinguish 6 classes of material:

- α.) Not affected by crater formation: Located far away from both craters.
- β.) Covered by formation of first crater, not affected by formation of second crater.
- γ.) Freshly exposed by formation of first crater. Not affected by formation of second crater.
- δ.) Freshly exposed by formation of first crater. Covered and thus shielded when second crater was formed.
- ε.) Not affected by formation of first crater, exposed by formation of second crater.
- ζ.) Not affected by formation of first crater, covered by second crater.

Similar to equations (26), (27) and (28) the amount of a stable spallation product formed may be expressed in a general form for each region

by an integral over the production rate:

$$C^{I_s}(x) = \int_{\tau_m}^{\tau_n} P^{I_s}(x,t) dt \quad (37)$$

If T_1 and T_2 are the ages of the two craters, determined by radioactive dating or from the local stratigraphy, then τ_m and τ_n are for the different regions:

region	τ_m	τ_n
α	$-\infty$	0
β	$-\infty$	T_1
γ	T_1	0
δ	T_1	T_2
ϵ	T_2	0
ζ	$-\infty$	T_2

Thus, the following equations hold for the concentrations of the different regions, independent of cosmic ray intensity and spectrum changes (corrected for differences in shielding and chemical composition)

$$\begin{aligned} C_{\alpha}^{I_s}(x) &= C_{\beta}^{I_s}(x) + C_{\gamma}^{I_s}(x) = C_{\epsilon}^{I_s}(x) + C_{\zeta}^{I_s}(x) \\ &= C_{\beta}^{I_s}(x) + C_{\delta}^{I_s}(x) + C_{\epsilon}^{I_s}(x) \end{aligned} \quad (38)$$

As in the case of the single crater, these relations can be used as an experimental check of our assumptions (no surface disturbances other than the formation of the two craters, no diffusion loss, no preirradiation).

The material of region δ has been irradiated only during a time interval in the past. This interval can be made short relative to the age of

the craters by choosing an appropriate set of craters. The average cosmic ray intensity $\overline{\phi(T_1, T_2)}$ in the time interval from T_1 to T_2 is then given by:

$$\overline{\phi(T_1, T_2)} = \frac{C_s^{I_s}(x)}{(T_2 - T_1) P^{I_s}(x, 0)} \quad (39)$$

Thus, from a single measurement on one sample from a two crater system it is possible to obtain the average cosmic ray intensity of a time interval in the past (assuming known crater ages). The result depends of course on the accuracy of the determination of the absolute age difference of the two craters. It is hard to say what precision in crater dating can be obtained, the limiting factor being diffusion loss. But it should be possible to reach an accuracy of the order of 50 m. y. even for craters as old as several 10^9 y. Thus a time interval of 500 m. y. could be determined with a 20% accuracy, making this method also sensitive to intensity variations far in the past (see 3.2.2.4).

3.2.2.3 Accumulating Surface Method

Let us assume for the present discussion that somewhere on the lunar surface a region can be found where new material is continuously accumulating, covering the old surface. GOLD (1955) suggested that the maria and certain craters are filled with dust which has accumulated over long periods of time. If we assume that the freshly accumulated material did not contain any spallation produced isotopes upon deposition, then the concentration in a depth x_0 can be calculated from:

$$C^{I_s}(x_0) = \int_0^{\infty} P^{I_s}(x(t), t) dt \quad (40)$$

Assuming a constant accumulation rate ε , then the present day concentration will be

$$C^{Is}(x_0) = \int_{-\frac{x_0}{\varepsilon}}^0 P^{Is}(x_0 + \varepsilon t, t) dt \quad (41)$$

Substituting the depth dependence found earlier for P^I (equation 20) and assuming that the spectrum of the cosmic radiation did not change with time and that the chemical composition of the deposited material is always the same we obtain:

$$C^{Is}(x_0) = P^{Is}(0,0) \int_{-\frac{x_0}{\varepsilon}}^0 \phi(t) \left[B_1 e^{-\mu_1(x_0 + \varepsilon t)} + B_2 e^{-\mu_2(x_0 + \varepsilon t)} \right] dt \quad (42)$$

$C^{Is}(x_0)$ can be determined experimentally by measuring the depth dependence of a stable spallation isotope, e.g. Ne^{21} , in a drill core. $P^I(0,0)$, the present surface production rate can be measured or calculated from the activity of a suitable radioactive spallation isotope in a sample (e.g. Na^{22}). B_1, B_2, μ_1, μ_2 are constants depending on the chemical composition. ε was assumed to be constant and can be obtained from the measurement of radioactive spallation isotopes (see 3.3). Thus (42) represents an integral equation for the time variation $\phi(t)$ of the cosmic radiation. If the chemical composition of the lunar material changes with depth a suitable correction can be applied.

This method makes the rather uncertain assumption of a constant deposition rate ε . It will probably be rather difficult to prove that this is indeed true in any specific case. Geological, mineralogical and petrographic arguments will have to be used. Comparison of the $\phi(t)$ obtained by this method from drill cores of different locations may also help to distinguish between changes in deposition rate and cosmic radiation. Changes

in the deposition rate may be locally different, whereas changes in $\phi(t)$ are the same on the whole lunar surface. (It is of course possible that also erosion rate fluctuations may be correlated on the whole lunar surface if they are induced by variations in the solar wind intensity, meteorite and dust influx rates, etc.)

3.2.2.4 Sensitivity of the Three Methods

In the preceding sections we have discussed three different methods for the detection of intensity variations of the cosmic radiation with lunar surface material as target. Of course, also the methods applied to meteorites could be used. Thus, for shorttime variations, (last few million years) the radioactive equilibrium of unstable spallation isotopes and for long time variations, the isotopic composition of spallation produced potassium might be useful (for a detailed description of these methods see: ARNOLD, HONDA and LAL 1961; GEISS, OESCHGER and SCHWARZ 1961; VOSHAGE 1962; GEISS 1963). We think that, regarding the lunar surface, the three methods introduced above are better and more sensitive.

To evaluate the relative sensitivity of the three methods, we assume three basic types of possible time variations of the cosmic ray intensity:

α.) short bursts

$$\begin{aligned} \phi(t) &= 1 && \text{all } t \\ \text{except} \quad \phi(t) &= \phi_1 && \tau_n + \Delta\tau_n < t < \tau_n \end{aligned} \quad (43)$$

β.) exponential increase or decrease with time

$$\phi(t) = e^{\lambda t} \quad -\infty < \lambda < \infty \quad (44)$$

γ.) oscillating intensity

$$\phi(t) = 1 + \phi_2 \sin \omega t \quad -\infty < \omega < \infty \quad (45)$$

Virtually no data are available to help us assume numerical values for the different constants. Measurements on meteorites indicate that the intensity was constant within a factor of two during the last few million years (ARNOLD, HONDA and LAL 1961) and may have slowly increased by about a factor of two during the last 10^9 years (VOSHAGE 1962). Thus, we have chosen the following parameters in equations (43), (44) and (45):

$$\begin{aligned} \alpha.) \quad \tau_n &= -1; \quad -10; \quad -100; \quad -1000 \text{ m.y.} \\ \Delta \tau_n &= -0.1; \quad -1; \quad -10; \quad -100 \text{ m.y.} \\ \phi_1 &= 2 \\ \beta.) \quad \lambda &= \pm 5 \times 10^{-10} \text{ y}^{-1} \\ \gamma.) \quad \omega &= 2 \times 10^{-8} \text{ y}^{-1}; \quad 2 \times 10^{-9} \text{ y}^{-1} \\ \phi_2 &= 1/2 \end{aligned}$$

Figure 6 shows the assumed cosmic ray intensity variations and figures 7, 8 and 9 the resulting concentration variations for the three different methods. It can be easily seen that the single crater method is the least sensitive one, especially for short bursts.

The two crater and accumulating surface methods show quite similar sensitivities. Both methods are, of course, closely related, the accumulating surface corresponding to a continuous two crater method. A constant accumulation rate and thus a constant absolute resolution was assumed for the accumulating surface method (figure 9), whereas for the two crater method a constant relative resolution ($(T_1 - T_2)/T_2 = \text{const.}$) was chosen (figure 8). With an accumulation rate of $10^{-6} \text{ cm y}^{-1}$, corresponding to a time constant of about 60 m.y., the accumulation method cannot any more detect the short bursts at -1 and -10 m.y.. Also the fast oscillations are already damped by about a factor of three. A higher accumulation rate would make the method more sensitive for such fast changes.

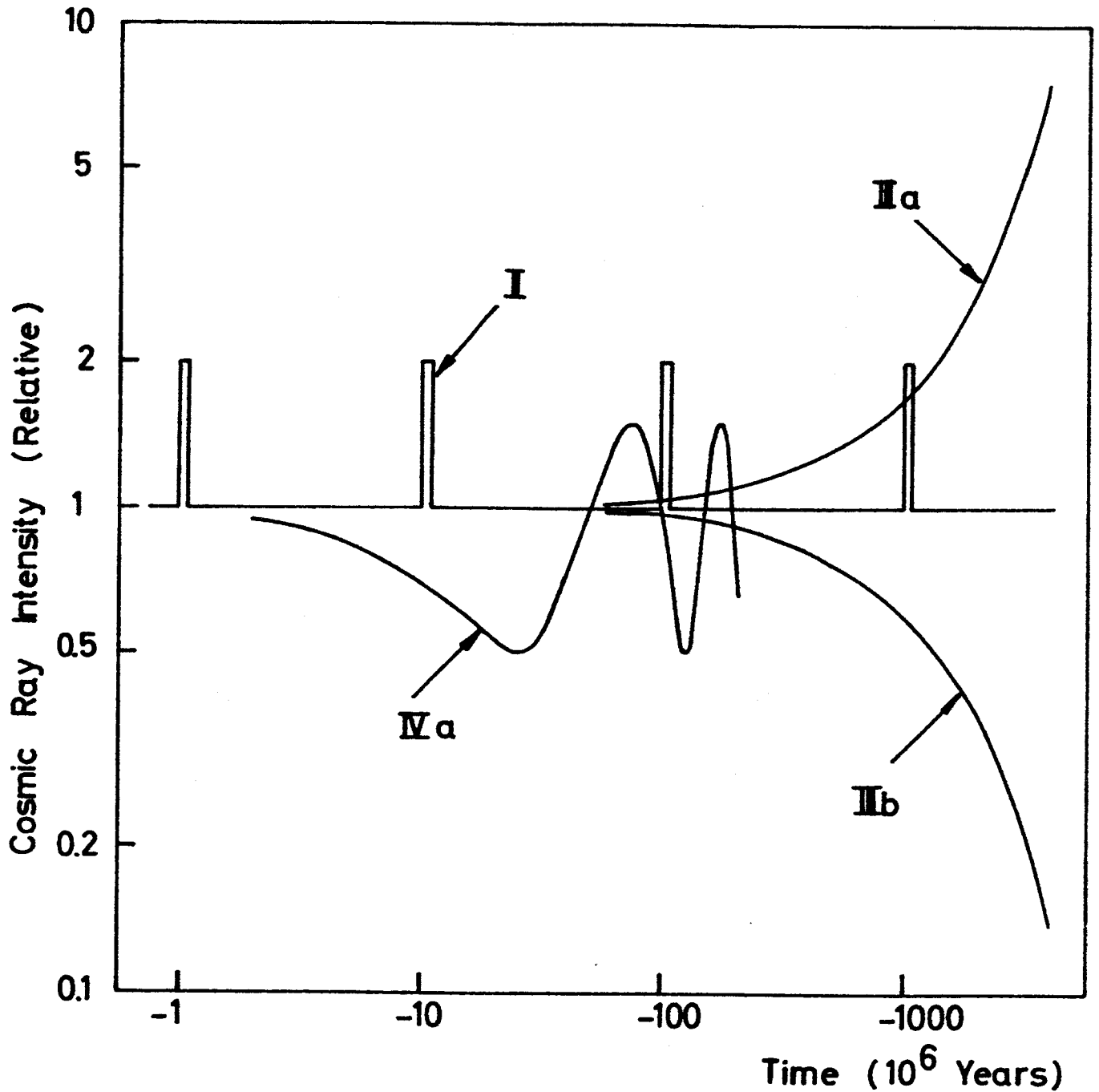


Figure 6. Assumed cosmic ray intensity variations for the evaluation of the sensitivities of the three detection methods. II : short bursts; IIIa : exponential decrease with time; IIIb : exponential increase with time; IVa : fast oscillation (slow oscillation, not shown, is similar to IVa, but shifted by order of magnitude). For constants used see 3. 2. 2. 4.

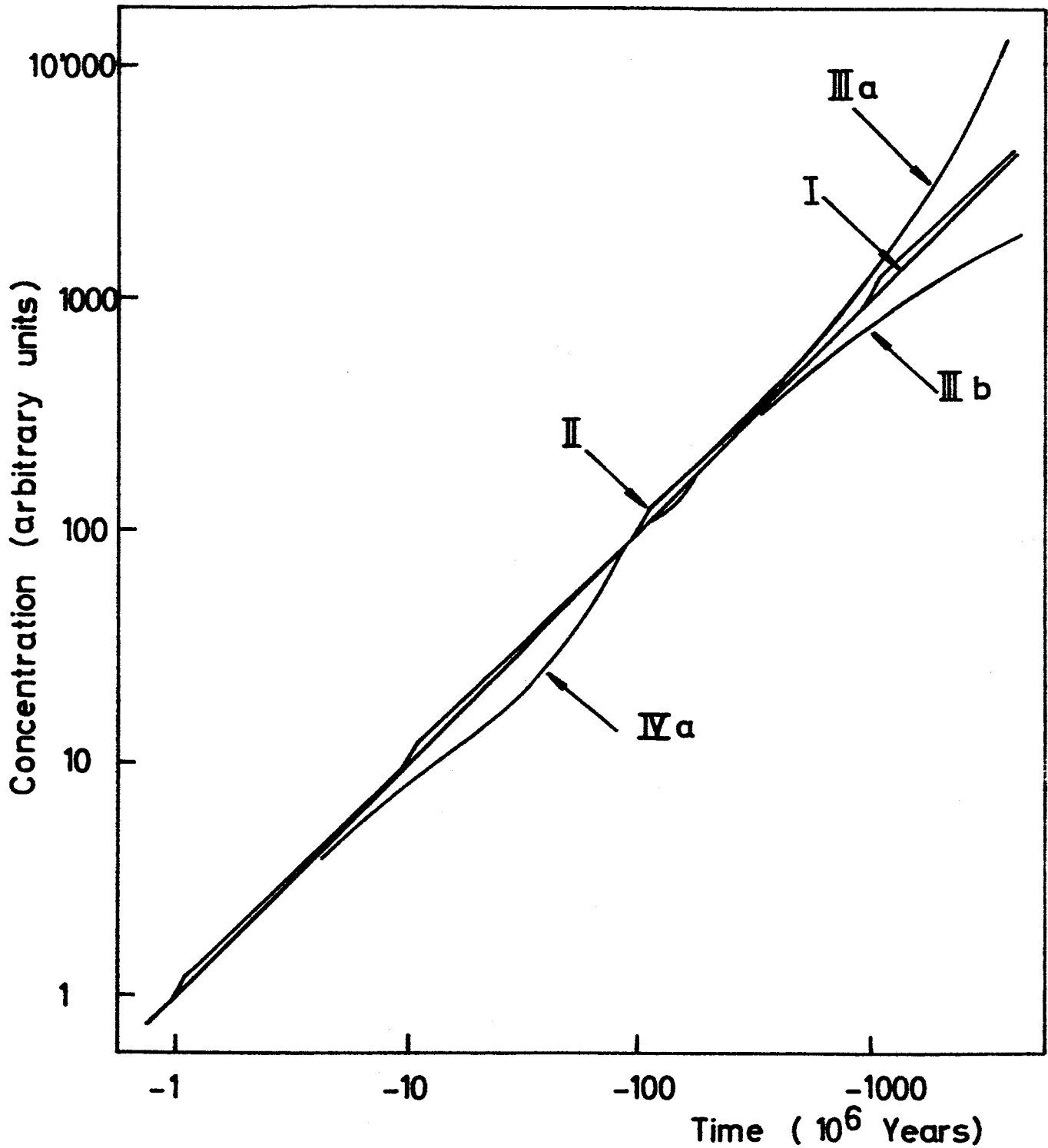


Figure 7. Spallation isotope concentration in single crater method resulting from different types of cosmic ray intensity variations. I : constant intensity; II : short bursts; IIIa : exponential decrease with time; IIIb : exponential increase with time; IVa : fast oscillation (slow oscillation, not shown, is similar to fast one but shifted by order of magnitude).

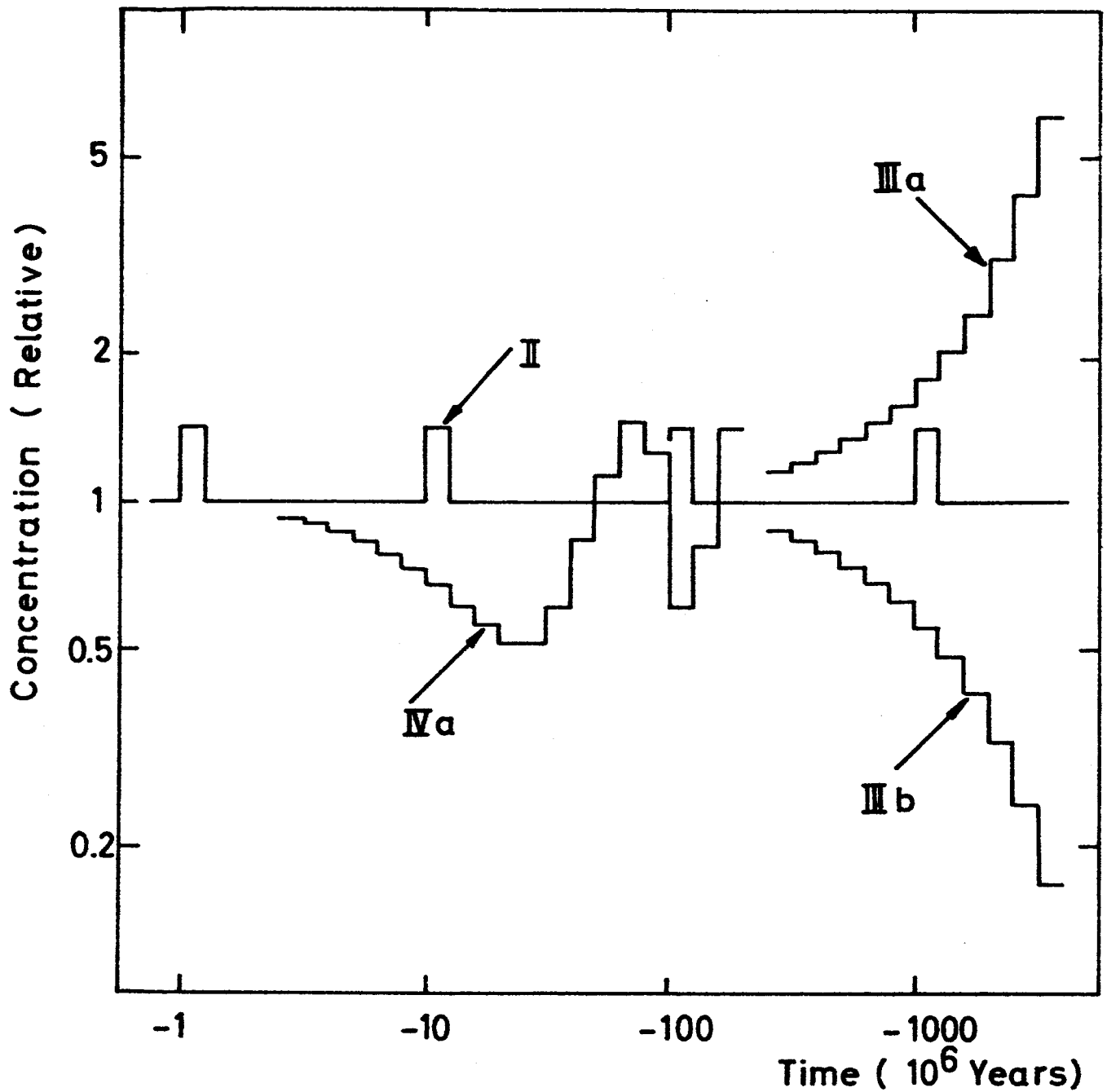


Figure 8. Average concentrations of spallation isotopes in two crater method produced by: II: short bursts; IIIa: exponential decrease with time; IIIb: exponential increase with time; IVa: fast oscillation (slow oscillation, not shown, is similar to fast one but shifted by order of magnitude). Assumed exposure interval: 25 per cent of age of second crater.

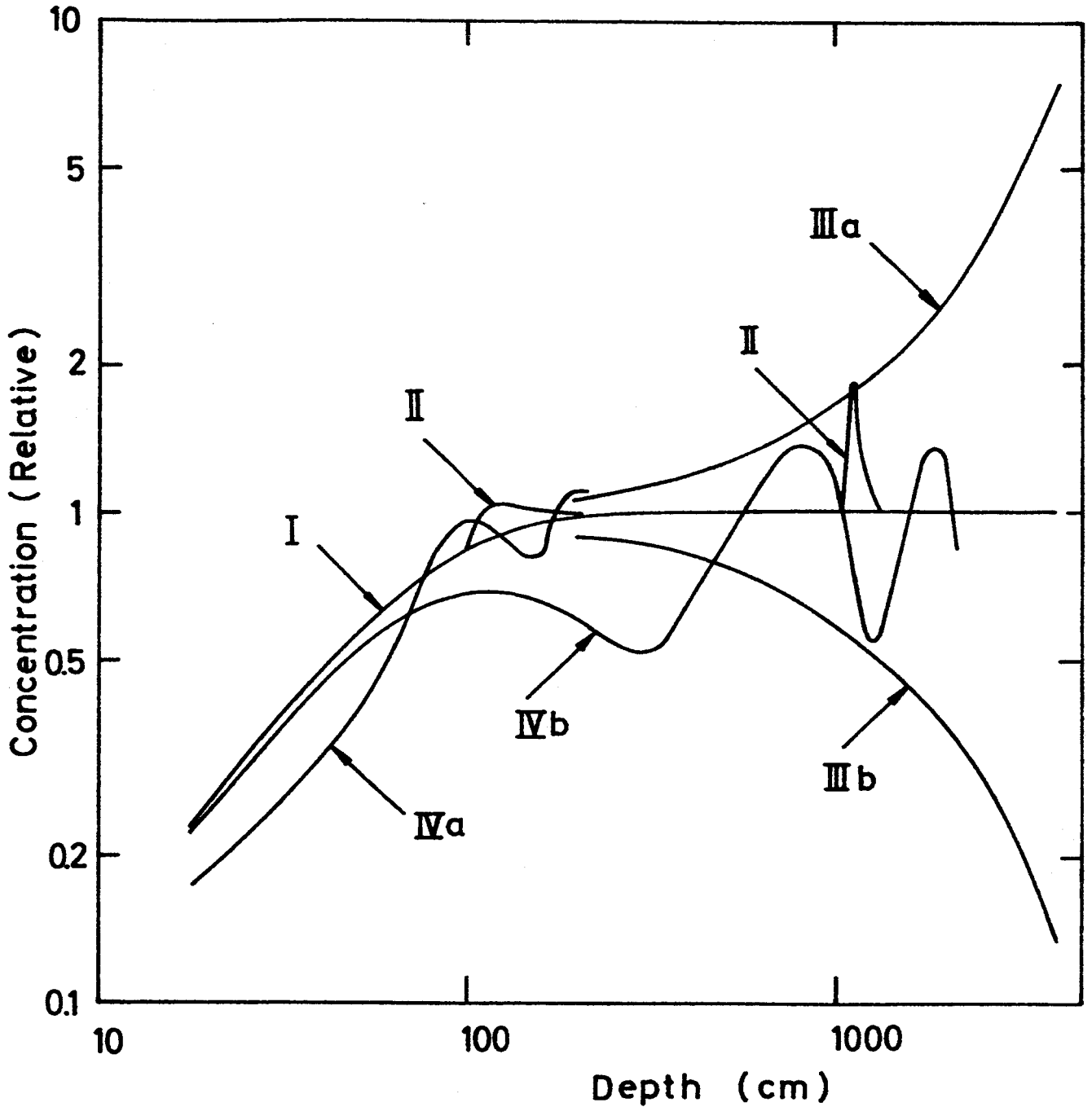


Figure 9. Depth dependence of spallation produced isotopes in accumulating surface method for different types of cosmic ray intensity variations. I : constant intensity; II: short bursts (only two shown); IIIa: exponential decrease with time; IIIb: exponential increase with time; IVa: fast oscillation; IVb: slow oscillation. Assumed accumulation rate $10^{-6} \text{ cm y}^{-1}$.

The sensitivity of the two methods is therefore about equal. Sampling for the accumulating surface method would be much easier, requiring only one drill core from a suitable location, whereas the two crater method would need samples from many two crater systems scattered probably over the whole lunar surface. However, the two crater method will be important as a check of the results obtained by the accumulating surface method, especially in deciding whether any variations found are indeed due to changes in the cosmic ray intensity, or only to changes in the accumulation rate.

3.2.3 Spectrum Changes

The production ratio of two different spallation isotopes with different mass losses, e. g. Ne^{21} and Ar^{38} in the spallation of iron, is very energy dependent. Also, the ratio of the production cross sections of one isotope from different elements, such as Ar^{38} from calcium or iron, is a function of the energy of the infalling particle. It has been recognized that the spectrum of the primary cosmic radiation could be deduced from the measurement of such ratios (EBERHARDT and HESS 1960; GEISS, OESCHGER and SCHWARZ 1961).

Two different approaches are possible for the calculation of the irradiation spectrum from the spallation isotope distribution. ARNOLD, HONDA and LAL (1961) have numerically integrated equation (15) and obtained a good agreement of the calculated production rates with the experimentally determined ones. This shows that their assumed irradiation spectrum is indeed the true one. The influence of spectral changes on the spallation isotope distribution is rather cumbersome to discuss because of the none analyticity of this method. GEISS, OESCHGER and SCHWARZ (1961) have given an analytical mathematical treatment of this problem. If the differential irradiation spectrum obeys a power law

$$g(E) dE = g_0 E^{-\alpha} dE \quad (46)$$

then the ΔA distribution of spallation products can be represented by

$$N(\Delta A) = C(\Delta A)^{-n} \quad (47)$$

with

$$C = \frac{3}{2} \frac{\sigma_0 A_0^{2/3} g_0 \Gamma(n)}{c^{n-1}} \quad (48)$$

where:

A_0	:	mass of target nucleus
ΔA	:	mass loss in spallation
σ_0	:	60 mb
$N(\Delta A)$:	total cross section for isobar ($A_0 - \Delta A$)
c	:	constant, depending on A_0

The observed spallation isotope distribution $N(\Delta A)$ in iron meteorites is in very good agreement with the one calculated from equation (47), at least in the range $11 < \Delta A < 35$. The irradiation spectrum is of course different from meteorite to meteorite, depending on shielding. The primary cosmic ray spectrum could be deduced, if such measurements were made on a very small iron meteorite.

It is obvious that this method will be very useful for measuring the cosmic ray spectrum in the past if applied to lunar material. The most suitable material will be that of region δ (see 3.2.2.2). The first crater formation exposes the material suddenly to the cosmic radiation. The second one will terminate the irradiation, probably without greatly disturbing the relative geometry of the irradiated material. The surface during the irradiation time, and thus the shielding of any sample, can be recognized from such surface effects as blackening (or other changes) by ultraviolet and particle radiation, by the presence of trapped rare gases, etc. A sample for a spectrum determination should of course be taken as close to the surface as possible.

The straightforward application of equation (47) requires that material with uniform atomic weight A_0 , such as iron meteorites or the metallic iron-nickel particles of chondrites, is available. Also oxides, e.g. iron oxide or titanium-oxide, or carbides, such as calcium carbide (UREY 1961), would be suitable, since oxygen or carbon will not contribute to the production of neon and argon.

If elements lighter than iron but heavier than neon are present, as e.g. in stone meteorites, it is not any more possible to apply equation (47). Of course, equation (47) can be summed over all elements present, but the method will be less sensitive to spectrum changes because most of the spallation isotopes will be produced mainly by lower energy particles from neighboring elements. Furthermore, equation (47) may not hold for small ΔA , requiring a numerical integration of equation (15).

3.2.4 Low Energy Particle History

Particles with energies below a few MeV are generally not able to induce nuclear reactions. Furthermore, because of ionization loss, their range is very limited. Even particles with energies of a few hundred MeV are stopped in a relatively small amount of shielding. Figure 10 gives the range-energy relation for protons in aluminum, which should be similar to the one for chondritic composition. Thus, only a sample with much less shielding than 70 g/cm^2 can give any information on the intensity or spectrum below an energy of 300 MeV. Very careful sampling and a good knowledge of the erosion history will be required to obtain any meaningful information for this energy range.

The very low energy particles, including the solar wind itself, will not induce any nuclear reactions and will simply be stopped and retained in the immediate surface layer of the moon. A certain amount of erosion by sputtering may result from this low energy bombardment of

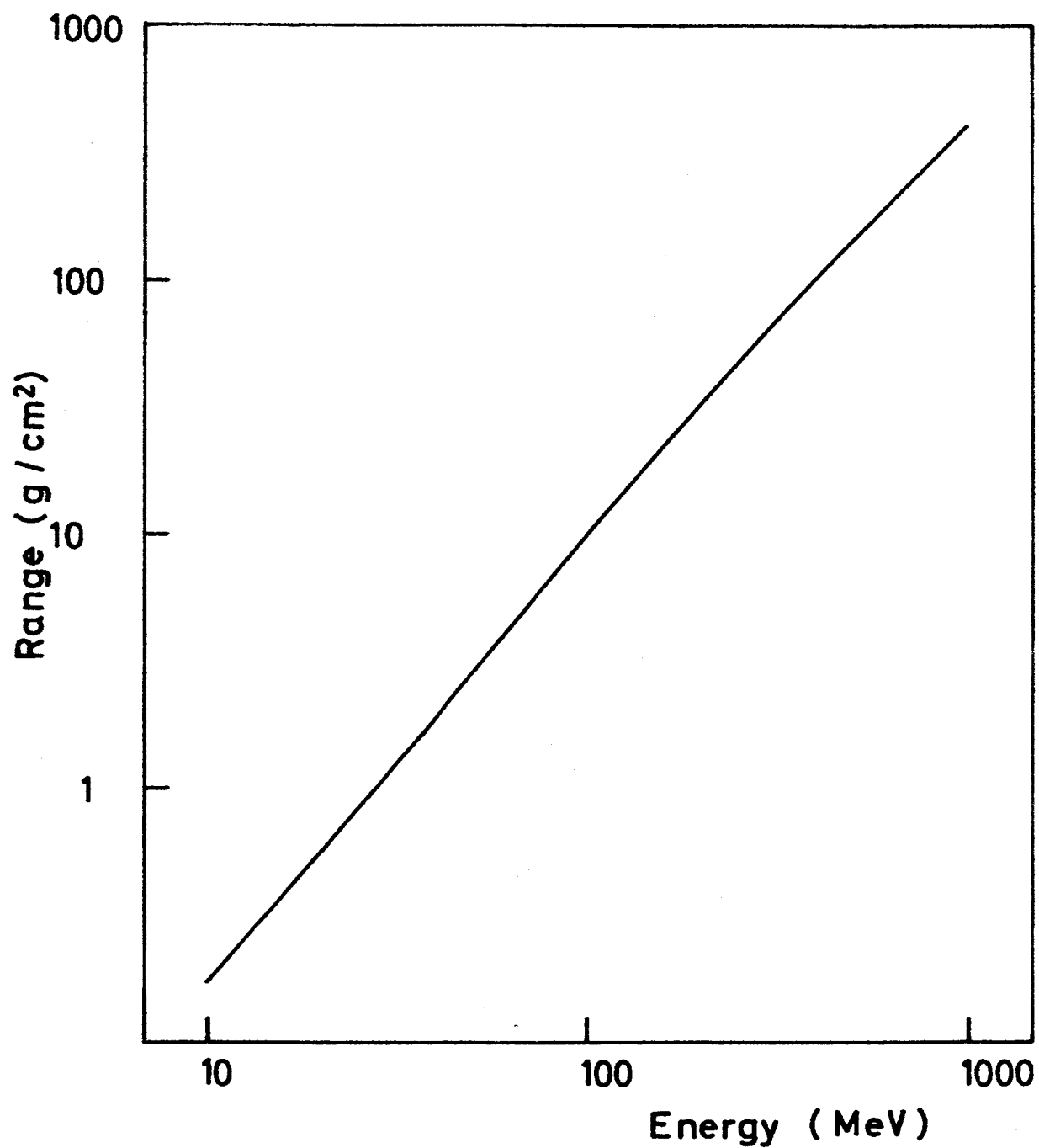


Figure 10. Range-energy relation of protons in aluminium (data from BETHE and ASHKIN 1953).

the surface. (see 3.3.1). The elemental abundance of the solar wind is most likely similar to the cosmic abundance. An irradiation time of one month with an average quiet sun solar wind intensity would already give a He^4 concentration of $2 \times 10^{-6} \text{ cc/cm}^2$. The corresponding amount of xenon, the least abundant of the rare gases, would be about $2 \times 10^{-15} \text{ cc/cm}^2$, a concentration measurable with material from 100 cm^2 of the surface.

That such trapping occurs is a well known phenomenon (ALMEN and BRUCE 1960). Surface concentrations of 10^{-3} cc/cm^2 for krypton-ions of 50 keV on aluminum have been obtained. The limiting concentrations are probably given by sputtering, and thus still higher concentrations should be possible for the lighter rare gases, which have much lower sputtering coefficients. Again, region δ and also a continuously accumulating surface would yield suitable material.

3.3 SURFACE EROSION

3.3.1 General

Four possible causes for surface erosion and turnover exist: impact cratering by meteorites; erosion by low energy particle radiation (solar wind) and electromagnetic radiation; turnover by volcanic and similar actions and erosion by temperature fluctuations. Nothing can be said at the present about the importance of the last factor. The observations of KOZYREV (1960) indicate that events similar to volcanic activity may indeed take place on the lunar surface, but nothing specific can yet be said on their frequency and importance.

Erosion by low energy particles will certainly take place on the lunar surface and it is possible to estimate roughly its magnitude. HEYMANN and FLUIT (1962) have measured the erosion rate of stone and iron meteorites by bombardment with 20 keV Ar^+ -ions, and found a value of

about 1.2 mg/coulomb. This value is in agreement with measurements of sputtering coefficients of iron and copper targets. Taking into account the much smaller sputtering coefficients of hydrogen and helium and assuming cosmic composition of the low energy particles one obtains an erosion rate of 0.03 mg/coulomb (HEYMANN and FLUIT 1962). The sputtering coefficient depends of course on the energy of the infalling particles. In the energy range between 5 keV and 70 keV the coefficient is about constant for light particles such as neon, nitrogen and argon. Below 5 keV it decreases, (ROL, FLUIT and KISTEMAKER 1960; ALMEN and BRUCE 1960). Thus, the erosion rate for the low energy solar wind particles would probably be not higher than 0.01 mg/coulomb. The maximal quiet sun solar wind particle flux of 10^9 particles/cm² sec would then yield an erosion rate of 5×10^{-5} mg/cm² y. In 10^9 years this would correspond to an erosion of 15 cm (with density of 3 g/cm³). The higher fluxes and energies observed during active sun periods will, because of their relatively short durations, not significantly increase this value. The same holds for events such as the recently observed luminescence of the lunar surface induced probably by medium energy particle irradiation (KOPAL and RACKHAM 1963).

The impact of meteorites and dust particles will lead to a constant turnover of the surface layer. Larger objects will produce craters big enough to be easily detected and thus their influence can be avoided by taking samples elsewhere. The magnitude of the turnover induced by smaller objects can be estimated from observed crater frequency distributions. DODD, SALISBURY and SMALLEY (1963) have shown that the crater frequency/crater size relation on the lunar surface can be approximated by a power law:

$$K(D) = A \frac{1}{D_0^{B-1}} D^B \quad (49)$$

where:

$K(D)$: number of craters with diameter
between D and $D + dD$
 D : crater diameter
 A and B : constants

DODD et al (1963) found values for B between -2.5 and -3.1 (except in the neighborhood of large craters where small secondary craters from ejected fragments are frequent and lead to values of $B = -4.8$).

The total surface covered by craters with diameters between D_1 and D_2 is

$$S(D_1, D_2) = \frac{\pi}{4} \frac{A}{D_0^{B-1}} \int_{D_1}^{D_2} D^{B+2} dD \quad (50)$$

With $B = -2.8$ we obtain:

$$S(D_1, D_2) = \frac{5\pi}{4} A D_0^{1.8} (D_2^{0.2} - D_1^{0.2}) \quad (51)$$

If we assume that the frequency size distribution holds also for smaller craters, then the total surface covered with craters smaller than D_2 will be

$$S(D_2) = \frac{5\pi}{4} A D_0^{1.8} D_2^{0.2} \quad (52)$$

With the highest A -value given and $D_2 = 10$ km one obtains a crater covered surface $S(D_2) = 0.1 \text{ km}^2 \text{ crater/km}^2$. Thus, only 10% of the whole surface would be covered by craters smaller than 10 km. The result depends of course strongly on B . If B would increase for smaller craters, then the whole surface would be crater covered. In this case the dust layer produced by the smaller impacts will, if no dust is removed by other processes, act as a shield of the underlying surface against further im-

pacts. Thus, the smaller impacts, even if they are frequent, will not necessarily change the irradiation geometry of the samples, provided they are at a certain minimum depth.

In the preceding chapters the assumption was often made that there was no surface erosion. The results obtained will of course change if erosion cannot be neglected. It therefore is necessary to have some criterion to decide whether positive or negative erosion of the surface has taken place or not. Erosion rate measurements will also be interesting for the interpretation of the history of the moon and the history of small particles in the solar system.

The geological setting of the sample will already offer some clues concerning its erosion history. But also the measurement of spallation products and rare gases will provide information. This will be discussed in the next three sections.

3.3.2 Stable Spallation Isotopes

The depth dependence of the concentration of a stable spallation isotope depends on the variation of shielding with time. This has been shown in detail in our discussion of the history of the cosmic radiation (see 3.2.2.3). A surface with a constant accumulation rate was discussed for the detection of cosmic ray intensity variations in the past.

If once the intensity function $\phi(t)$ is known, then the depth dependence will be useful for measuring the erosion history of any particular area of the lunar surface. The concentration of a stable spallation isotope at depth x_0 is then

$$C^{I_s}(x_0, 0) = P_{(0,0)}^{I_s} \int_{-\tau_0}^0 \phi(t) \left[B_1 e^{-\mu_1 x(t)} + B_2 e^{-\mu_2 x(t)} \right] dt \quad (53)$$

$x(t)$ is the only unknown in this integral equation if the depth dependence of $C^{Is}(x_0, 0)$ has been determined experimentally. This method is more sensitive to accumulation than to erosion.

If for any particular surface the beginning of the exposure to cosmic radiation is known from an independent measurement, e.g. the age of an ejecta blanket from the age of its crater, then this age can be compared with the radiation age obtained by measuring the absolute amount of a stable spallation isotope. These ages should be the same if no erosion or accumulation has occurred. If they do not agree the average erosion rate during the exposure time of the surface can be calculated. This method is not only sensitive to accumulation but also to erosion.

3.3.3 Trapped Gases

As discussed before (3.2.4) the immediate surface layer of the moon is exposed to the low energy particles of the solar wind. These particles will be trapped in the grains of the surface layer.

The solar wind most likely has an elemental abundance similar to the cosmic abundance and thus will contain also the rare gases. Any mineral grain which was once on the immediate surface will therefore contain large amounts of trapped gases. Thus, the concentration of these gases may be an indicator for the surface-residence time of a mineral grain.

3.3.4 Radioactive Spallation Isotopes

Radioactive spallation isotopes will be present in a sample which has been irradiated up to the moment of collection. The activity of all these isotopes will be in equilibrium with the production rate if the cosmic ray intensity and the shielding have not changed during the last few half-lives. If the shielding has changed recently or continuously, the longer lived isotopes will not yet be in equilibrium with the present day production rate. Thus, from a measurement of two radioactive isotopes with different half-lives, such as H^3 and Cl^{36} , the erosion rate could be obtained.

Drawbacks of this method are the relatively short half-life ranges of radioactive spallation products ($0 < T_{1/2} < 2,5$ m. y., except K^{40}), limiting this method to the recent erosion history. Furthermore, much larger samples are required for a determination of radioactive spallation isotopes than for stable rare gases. If suitable samples can be found (low primordial potassium content, high "heavy" element ($A > 40$) concentrations) also K^{40} could be used as a radioactive spallation isotope.

3.4 DIFFUSION LOSS

Up to now we have always made rather specific assumptions on diffusion losses. Either the diffusion loss was complete, so that the material lost its memory of previous irradiations etc., or no diffusion loss at all was assumed. The results obtained on meteorites have shown that deviations from such a simple diffusion history sometimes do occur. Diffusion loss may be especially important for the immediate surface material of the moon because of the high temperatures reached during the lunar day. We know from infrared measurements that the lunar surface reaches a maximal average temperature of $120^{\circ}C$. (For a review of temperature measurements of the lunar surface see SINTON 1962). For

this temperature FECHTIG, GENTNER and LAEMMERZAHN (1963) found average D/a^2 - values in the range of 10^{-12} to $10^{-18} \text{ sec}^{-1}$ for chondrites. The higher values generally hold for helium, the medium for neon and the lower values for argon. Thus, depending on the residence time of an individual grain on the lunar surface, diffusion losses may indeed be important.

The lunar subsurface temperature is much lower. Measurements of the centimeter and millimeter wave emission of the lunar surface give maximal temperatures of -20°C . (Not the average but the maximal temperature will determine the amount of diffusion loss, because D depends exponentially on the temperature). Extrapolating the data of FECHTIG et al (1963) to such low temperatures leads to D/a^2 values of $10^{-23} \text{ sec}^{-1}$ and lower. Thus, diffusion losses should be negligible for subsurface samples, provided no additional heating by volcanic action, etc. has taken place.

Nevertheless, it will be necessary to check experimentally a sample's diffusion loss history. (With known diffusion coefficients and activation energies it will be possible to reinterpret the diffusion loss history as the temperature history of the sample). The following methods could be applied:

- a.) Comparison of results from methods involving different rare gases for measuring the same quantity. Diffusion coefficients of the individual rare gases are different, the heavier generally having much lower values. Thus, for instance the agreement of the K-Ar and U/Th-He ages would exclude any erratic diffusion losses. The spallation produced isotopes He^3 , Ne^{21} and Ar^{38} may be used independently for the determination of the total irradiation dose of the sample and their agreement will exclude diffusion losses. In extreme cases, where all the light rare gases show diffusion losses, krypton and xenon may not be affected.

- β.) Measurements on different minerals or components of the same sample should give the same results. Differences could of course also be due to different histories of the minerals prior to their accumulation.
- γ.) Grain size dependence of rare gas concentrations (corrected for chemical differences, etc.). This method has already been discussed earlier (see 2.2.3.2). Differences may again be due to different histories of the grains before accumulation.
- δ.) Comparison of results obtained on different samples. Different samples, dating the same event, such as glasses from various locations in a crater should give the same results. The history of the cosmic radiation deduced from different samples should be the same, etc.

These four methods will not only give information on the diffusion history, but will also be useful as checks on the other "boundary conditions" made in our earlier discussions of the applications of rare gas measurements to specific problems.

3.5 CONCLUDING REMARKS

We have discussed several possible applications of rare gas measurements to lunar samples. These are by no means the only possible ones, but in our opinion constitute the more important ones. They show the value of rare gas measurements for investigating the history of the moon, the history of the solar system and the history of the particle radiation field in the solar system.

In many cases we have assumed that material of selected lunar areas or of a specific history is available. This was done to show what material is most suitable for solving any specific problem and to serve as a guide for sampling. Moreover, rare gas measurements will still be extremely useful, even if the available material does not exactly correspond to these specifications. One should remember, that the sampling in meteorites is completely random and afflicted with many unknown parameters. Nevertheless, rare gas measurements have been the most successful contributors to our knowledge of the history of meteorites and their parent bodies.

Also, we can expect that future research on meteorites will lead to a still better understanding of many problems and to a development of even more sophisticated sample preparation and rare gas measurement techniques before lunar samples are available. Thus, we should consider the applications mentioned in this paper only as "lower limits" of the possibilities of rare gas measurements on lunar samples.

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